



Ammonia variability and trends from globally distributed FTIR measurements and model simulations

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Abstract. Ammonia (NH₃) is an important constituent in the global nitrogen cycle, present in both urban and remote environments. It is a source of reactive nitrogen and a precursor for particulate matter, thereby affecting atmospheric chemistry and radiative forcing. This work presents the seasonal and diurnal variability, along with long-term trends, of atmospheric NH₃ total columns retrieved from Fourier transform infrared (FTIR) spectroscopic solar absorption measurements at 22 ground-based sites, globally distributed from 45°S to 80°N. Comparisons are made with simulations from the GEOS-Chem High Performance (GCHP) chemical transport model and the Tropospheric Chemistry Reanalysis (TCR-2) NH₃ product. The mean NH₃ total columns from the FTIR time series ranged from 0.12×10^{15} to 19.20×10^{15} molecules cm⁻², with the smallest columns found at the Arctic and high-altitude sites, and the largest in urban areas. Significant enhancements were attributed to



biomass burning, and NH_3 emissions from volcanic eruptions were detected at the Izana site. The seasonal patterns are similar across most sites, with maxima mainly related to the volatilization of NH_3 due to higher temperatures. The diurnal variability differs significantly and depends on the characteristics of each site and local sources. Most sites have positive trends in the total column, with a mean value (and 95% confidence interval) for all sites of 3.82 (3.29–4.35)% for the FTIR measurements, 3.66 (3.35–3.97)% for GCHP, and 6.49 (2.00–10.98)% for TCR-2. GCHP exhibited a better general agreement with the FTIR observations than TCR-2; potential reasons for this are explored, including a sensitivity test on the emissions used.

15 1 Introduction

The nitrogen cycle has changed significantly since the introduction of the industrial fixation of nitrogen (Battye et al., 2017). With the growing world population and demand for food, fertilizer application has been increasing, especially since 1960 with the introduction of synthetic nitrogen fertilizers; it is estimated that currently, the anthropogenic flux of fixed or reactive nitrogen to the biosphere is roughly equivalent to the total flux from natural sources (Battye et al., 2017). Part of this nitrogen is lost to the environment in the form of emissions of nitrogen compounds, like ammonia (NH_3) (Paulot et al., 2014; Battye et al., 2017) for which emissions might reach 130 Tg $\text{NH}_3\text{-N yr}^{-1}$ by 2100 (Sutton et al., 2013). Emissions of reduced N to the atmosphere are a key driver for atmospheric composition and chemistry in the twenty-first century (Fowler et al., 2013).

In the atmosphere, NH_3 is the most abundant alkaline compound. It is involved in several chemical reactions, such as the neutralization of atmospheric acidity and the formation of particulate matter, with strong consequences for the environment and human health (Behera et al., 2013). The main sources of NH_3 are related to agricultural activities (Sutton et al., 2008, 2013); for example, according to an estimate made by Paulot et al. (2014), out of the total global emissions of 54 Tg $\text{NH}_3\text{-N yr}^{-1}$, 34 Tg $\text{NH}_3\text{-N yr}^{-1}$ come from manure and fertilizer applications, with the remainder related to other natural and anthropogenic sources such as open fires or industries. Once in the atmosphere, the primary chemical pathways are wet and dry deposition and conversion to particulate form (Behera et al., 2013; Sutton et al., 2013; Khan et al., 2020).

NH_3 emissions contribute significantly to $\text{PM}_{2.5}$ formation by its conversion to ammonium salts, which act as precursors and can account for up to 50% of particulate matter mass (Behera et al., 2013), a key factor in premature human mortality (Paulot et al., 2014; Giannakis et al., 2019). Despite the importance of atmospheric NH_3 , most countries around the world do not have regulations to directly reduce NH_3 emissions, compared with the efforts to reduce NO_x and SO_2 emissions (Behera et al., 2013). Control of NO_x emissions contributed to the reduction in deposition of oxidized nitrogen (NO_y = the sum of all oxidized nitrogen species except N_2O). However, deposition of reduced nitrogen (NH_x) such as NH_3 increased (Tan et al., 2020). In addition, future increases in emissions are expected due to the potential use of NH_3 as a hydrogen carrier for the production of renewable energy, specifically using industrially-produced NH_3 with no or almost no carbon dioxide (green ammonia) for which production has increased in popularity in recent years (Olabi et al., 2023).

NH_3 has a short lifetime in the atmosphere, on the order of hours to a few days (Dammers et al., 2016, 2019; Nair and Yu, 2020; Evangelidou et al., 2021). NH_3 emissions and deposition strongly depend on environmental conditions; temperature and moisture play a crucial role in determining NH_3 concentrations on diurnal to annual scales (Sutton et al., 2013). NH_3 exhibits



a strong temporal and spatial variability that ranges over three orders of magnitude near the surface (Shephard et al., 2011). Previous studies, mainly using satellite observations, have shown a clear NH_3 seasonal cycle with temperature dependence in several sites around the world, with the seasonality driven by management differences and peaks during fertilizer of manure applications where applicable, and the interannual variability is attributed to meteorological conditions and agricultural constraints (Sutton et al., 2013; Paulot et al., 2014; Viatte et al., 2020).

Van Damme et al. (2015) used satellite data to show the worldwide spatiotemporal variability of atmospheric NH_3 using morning and evening measurements by the Infrared Atmospheric Sounding Interferometer (IASI) between 2008 and 2013. For most regions, defined seasonal cycles were reported, with the greatest concentrations found in Asia. The NH_3 columns vary widely around the world. For example, for Central America, the highest columns were during April and March and for South America during September and October, while Europe and North America showed higher columns during the spring season with columns around $1.5 - 2 \times 10^{16}$ molec. cm^{-2} . However, IASI measurements tend to underestimate NH_3 total columns compared to ground-based FTIR data (Dammers et al., 2016), and with limited measurements (Van Damme et al., 2015), it is not possible to determine the diurnal variability. In addition, NH_3 from biomass burning emissions contributes to seasonal variability over certain regions, for example, Central South America, as described by Whitburn et al. (2015), where the interannual variability of Fire Radiative Power (FRP) represented the intensity of NH_3 and CO enhancements from fire events between August and October.

Previous studies have investigated NH_3 trends in different regions using satellite or ground-based data. In North America, using surface data from monitoring stations distributed across Canadian and American urban, rural, and remote sites, positive trends in NH_3 mixing ratios between 20% and 50% from 2008 to 2015 at urban and agricultural sites were reported, mainly attributed to changes in the $\text{NH}_3 - \text{NH}_4^+$ partitioning (Yao and Zhang, 2016). In addition, more recent positive trends in reduced N in the USA have also been reported by Benish et al. (2022). More studies such as Yamanouchi et al. (2021) and Herrera et al. (2022) explored NH_3 over Toronto and Mexico City respectively using solar Fourier transform infrared spectroscopy (FTIR), also finding positive trends. In China, Chen et al. (2023) reported that according to several inventories NH_3 emissions from different anthropogenic sectors have doubled since 1980. In terms of global approaches, Van Damme et al. (2021) used ten years of global satellite data to derive trends of NH_3 total columns, reporting a worldwide increase of $12.8 \pm 1.3\%$ between 2008 and 2018, with increases over east Asia, western and central Africa, western and southern Europe, and North America; mainly attributed to anthropogenic factors affecting the lifetime of atmospheric NH_3 . Warner et al. (2017) reported positive trends of NH_3 over several major agricultural areas around the world, with drivers mainly related to changes in fertilizers use, control of sulphur and nitrogen oxide emissions, and increasing temperatures.

This work uses NH_3 total columns from ground-based FTIR measurements, and NH_3 simulations, from the the GEOS-Chem High Performance (GCHP) global chemical transport model (CTM) and the Tropospheric Chemistry Reanalysis product (TCR-2), to examine the variability and trends of atmospheric NH_3 at 22 sites globally distributed, in particular the diurnal variability that has been poorly explored in previous studies. The paper is organized as follows. Section 2 describes the datasets and methods, starting with the FTIR site characteristics and retrieval strategy, followed by the model, reanalysis product, and trend analysis equations. Section 3 presents the FTIR measurements and the variabilities and trends. Section 4 describes



the comparison between the FTIR and modeled NH_3 products at each site. Finally, Section 5 summarizes the most relevant conclusions of this work.

2 Datasets and Methods

80 2.1 Ground-based FTIR NH_3 Measurements

2.1.1 FTIR Sites

Most of the sites included in this study are affiliated with the Network for the Detection of Atmospheric Composition Change (NDACC) (De Mazière et al., 2018), contributing data from high-resolution FTIR spectrometers. Most sites use the Bruker Optik GmbH IFS 120/125 HR spectrometer and record solar absorption spectra with a typical spectral resolution of 0.0035
85 cm^{-1} , equipped with either KBr or CaF_2 beamsplitters, and HgCdTe and InSb detectors, with three exceptions. The Toronto instrument is a high resolution ABB Bomen DA8, that has been providing measurements since mid-2002. The Mexico City instrument is a low-resolution Bruker Optik GmbH Vertex 80, which was included in this study due to the relevance of the site, as it is located in the south of Mexico City, one of the largest metropolitan areas in the world, and the largest in North America. The Los Angeles instrument is the MkIV high-resolution FTIR spectrometer, which has been performing ground-
90 based observations since the 1980s, mainly in California (Toon, 1991; Sen et al., 1996; Toon et al., 2018).

Table 1 presents the main characteristics of each of the 22 FTIR sites. The primary NH_3 sources are based on the emissions inventories used for the GCHP simulations (described in Section 2.2.1) and are sorted by their contribution relevance for each site. Eureka, Thule, and Ny Alesund are Arctic sites where NH_3 sources are limited. Zugspitze, Jungfraujoch, Izana, Altimoni, and Reunion Mado are high-altitude stations, with generally smaller NH_3 columns. St. Petersburg, Toronto, Los
95 Angeles, Hefei, and Mexico City are sites located in heavily urban areas with populations of millions. Bremen, Boulder, Tsukuba, Paramaribo, Porto Velho and Wollongong are located in less urbanized areas, while Garmisch, Reunion St. Denis, and Lauder are more remote sites. Data from the NDACC FTIR instruments have been used to accurately document trends of many stratospheric and tropospheric gases (De Mazière et al., 2018).

2.1.2 FTIR NH_3 Retrieval Strategy

100 The retrieved NH_3 total columns presented in this work were mainly obtained following the strategy recommended in Dammers et al. (2015), which is briefly described in the following paragraph. For most sites, the solar FTIR spectra were analyzed using the SFIT4 retrieval algorithm (<https://wiki.ucar.edu/display/sfit4/>) version 0.9.4.4 to obtain the NH_3 total columns, except for Garmisch, Zugspitze and Izana, which used PROFFIT9 (Hase et al., 2004), and Los Angeles which used GFIT (Roche et al., 2021; Zeng et al., 2021; Laughner et al., 2024).

105 The SFIT4 retrieval uses two microwindows (MW) both in the ν_2 vibrational band of NH_3 : MW1 (930.32 to 931.32 cm^{-1}) and MW2 (966.97 to 967.675 cm^{-1}). For some sites, wider MW1 (929.40 to 931.4 cm^{-1}) and MW2 (962.10 to 970.0 cm^{-1}) or a third MW in between them (MW3, 950.2 to 952.0 cm^{-1}) were used, especially for sites with small ambient NH_3 concentra-



Table 1. FTIR sites used in this study. The primary NH₃ sources are from the emissions inventories used in the GCHP simulations.

Station	Latitude	Longitude	Elevation m.a.s.l.	Measurement Period	Primary NH ₃ Sources
Eureka	80.05°N	86.4°W	610	2006–2020	Anthropogenic, natural, biomass burning, seabirds
Ny Alesund	78.92°N	11.93°E	20	2017–2019	Natural
Thule	76.53°N	68.74°W	225	1999–2022	Seabirds, biomass burning
St. Petersburg	59.90°N	29.80°E	20	2010–2022	Anthropogenic, natural, biomass burning
Bremen	53.10°N	8.80°E	27	2004–2018	Anthropogenic, natural
Garmisch	47.48°N	11.06°E	743	2004–2007	Anthropogenic, natural
Zugspitze	47.42°N	10.98°E	2694	1995–2023	Anthropogenic, natural
Jungfraujoch	46.55°N	7.98°E	3580	2008–2023	Anthropogenic, natural, biomass burning
Toronto	43.66°N	79.40°W	174	2002–2023	Anthropogenic, natural, biomass burning
Boulder	39.99°N	105.26°W	1634	2010–2022	Anthropogenic, natural, biomass burning
Tsukuba	36.05°N	140.13°E	31	2014–2022	Anthropogenic, natural, biomass burning
Los Angeles	34.20°N	118.17°W	345	1985–2024	Anthropogenic, natural, biomass burning
Hefei	31.90°N	117.17°E	30	2017–2022	Anthropogenic, natural, biomass burning
Izana	28.30°N	16.48°W	2367	2000–2022	Anthropogenic, natural, biomass burning
Mexico City	19.32°N	99.17°W	2280	2012–2022	Anthropogenic, natural, biomass burning
Altzomoni	19.11°N	98.65°W	3985	2012–2022	Anthropogenic, natural, biomass burning
Paramaribo	5.81°N	55.21°W	23	2018–2019	Natural, anthropogenic, biomass burning
Porto Velho	8.77°S	63.87°W	87	2019	Biomass burning, anthropogenic, natural
Reunion St. Denis	20.90°S	55.50°E	85	2004–2011	Anthropogenic, natural
Reunion Maito	21.10°S	55.40°E	2155	2013–2023	Anthropogenic, natural
Wollongong	34.41°S	150.88°E	30	2007–2022	Anthropogenic, natural, biomass burning
Lauder	45.04°S	169.68°W	370	1996–2022	Anthropogenic, natural, biomass burning

tions. The retrievals included the interfering species H₂O, CO₂, O₃, CO, HNO₃ and N₂O for all stations, and temperature and pressure profiles obtained from the USA National Centers for Environmental Prediction (NCEP). Spectroscopic parameters were obtained from the high-resolution transmission molecular absorption database HITRAN 2008 (Rothman et al., 2009), except for St. Petersburg, which used HITRAN 2012 (Rothman et al., 2013). The a priori profiles for the interfering gas were obtained from 40-year averages of simulations from the Whole Atmosphere Community Climate Model (WACCM) (Eyring et al., 2007; Marsh et al., 2013). For NH₃, the profiles from WACCM are only representative of remote regions; for this reason most of the sites used scaled a priori profiles derived from five years of averaged NH₃ simulations from the global chemical transport model GEOS-Chem v11. Constructed and scaled a priori profiles from GEOS-Chem simulations have been used previously for the retrieval of trace gases with good results (Shephard et al., 2011; Shephard and Cady-Pereira, 2015; Bader et al., 2017; Herrera et al., 2022), and scale factors are empirically calculated based on the convergence of the retrieval. The



average degrees of freedom for signal (DOFS) for all sites was 1.07 ± 0.15 , indicating that the retrieval contains approximately one independent piece of information. The total error on the NH_3 total columns was calculated as the square root sum of the squares of all systematic and statistical errors (García et al., 2021), ranging between 0.1 and 2.0 with an average value for all sites of $0.93 \pm 1.30 \times 10^{15}$ molec. cm^{-2} .

2.2 Simulations of NH_3

2.2.1 GEOS-Chem

GEOS-Chem is a global 3-D model CTM (<http://geos-chem.org>) that is driven by assimilated meteorological fields from the NASA Global Modeling and Assimilation Office (GMAO). For this work, the high-performance version of GEOS-Chem was employed (Eastham et al., 2018; Martin et al., 2022). This version of GEOS-Chem runs on the native Goddard Earth Observing System cubed-sphere model grid. The analysis here uses the GCHP version 14.1.1 simulation from 2003 to 2021 driven by Modern-Era Retrospective Analysis for Research and Applications, version 2 (MERRA-2) (Gelaro et al., 2017), meteorological fields at C48 resolution, which is comparable to a resolution of $2^\circ \times 2.5^\circ$ (Bindle et al., 2021). The vertical resolution of the model consists of 72 levels from the surface to 0.01 hPa (approximately 80 km). Anthropogenic emissions are from the Community Emissions Data System (CEDS) v2 (Hoesly et al., 2018), biomass burning emissions from the Global Fire Assimilation System (GFAS) (Kaiser et al., 2012) v1.2, and natural emissions from the Global Emissions Inventory Activity (GEIA) (Graedel et al., 1993).

The analysis presented here uses a full-chemistry simulation that covers the period from 1 January 2003 to 31 December 2021. The model spin-up was performed for the full year of 2002 to provide the initial conditions for the simulation. The model transport and chemical operator time steps were selected for 10 and 20 minutes, respectively, as recommended by Philip et al. (2016) to minimize simulation errors. The modeled fields were archived at a temporal resolution of two hours for comparison with the FTIR observations. More details and applications of this long-term GCHP simulation can be found in Wizenberg et al. (2024). In addition to the 2003-2021 GCHP run, a one-month simulation was performed to explore the effect of changing the global emissions, by using the Hemispheric Transport of Air Pollution (HTAP) v3 emissions (Janssens-Maenhout et al., 2015), instead of CEDSv2, maintaining the same model configuration. The results of this simulation are discussed in Section 4.3.

2.2.2 TCR-2

The Tropospheric Chemical Reanalysis product (Miyazaki et al., 2020a) is a reanalysis product that consists of a global CTM and a data assimilation technique to optimize chemical concentrations and emissions of various chemical species. The TCR-2 ozone product is very well known and has been compared with several ozone reanalysis products, showing improved agreement with independent ground and ozone-sonde observations (Huijnen et al., 2020). In this work, the TCR-2 NH_3 product was evaluated. The reanalysis was produced using the MIROC-CHASER CTM (Watanabe et al., 2011) and an ensemble Kalman filter (EnKF) within the Multi-mOdel Multi-cOnstituent Chemical data assimilation (MOMO-Chem) (Miyazaki et al., 2020b) to assimilate observations of O_3 , CO , NO_2 , HNO_3 and SO_2 from multiple satellite instruments. The data assimilation system



150 simultaneously updates tropospheric emissions and chemical concentrations of 35 chemical species described in Miyazaki et al. (2012, 2015, 2020a). The representation of tropospheric chemistry, including NO_x and SO_2 emissions, is improved by assimilating NO_2 measurements from the Ozone Monitoring Instrument (OMI), the Global Ozone Monitoring Experiment (GOME-2), and the Scanning Imaging Absorption Spectrometer for Atmospheric Cartography (SCIAMACHY), and SO_2 from OMI.

155 Although NH_3 measurements were not directly assimilated in this product, the assimilation of NO_x and SO_2 can influence the NH_3 analysis. For instance, an analysis of the COVID-19 lockdown period showed that while NO_x and SO_2 emission reductions directly decreased nitrate and sulfate aerosols, they also led to a decrease in ammonium aerosols, even though NH_3 emissions remained unchanged in this analysis. This effect can be attributed to changes in NH_3 lifetime (Sekiya et al., 2023).

The TCR-2 product used in this work has a horizontal resolution of $1.1^\circ \times 1.1^\circ$ and consists of 32 vertical levels from the surface to 4.4 hPa. The TCR-2 fields were archived at a temporal resolution of 2 hours between 2005 and 2018. A priori anthropogenic emissions used in the reanalysis are from the HTAPv2 emissions (Janssens-Maenhout et al., 2015), biomass burning emissions from GFED v4 (Randerson et al., 2017), and soil emissions from GEIA (Graedel et al., 1993). The HTAP emission inventory has a $0.1^\circ \times 0.1^\circ$ resolution between 2000 and 2018 and is used to study trends and transport of air pollutants.

2.3 Trend Analysis

165 To derive long-term trends for the time series, we used a linear trend model with fitted Fourier series to account for seasonality, and bootstrap resampling methods to derive the 95% confidence intervals following the methodology of Friedrich et al. (2020). The trend model is described as follows:

$$y_t = d_t + s_t + u_t \quad (1)$$

where y_t are NH_3 measurements at time t , d_t is the unknown trend, s_t is the intra-annual seasonal pattern, and u_t is the error term. s_t is described by Fourier terms given by Equation 2.

$$s_t = \sum_{j=1}^S a_j \cos(2j\pi t) + b_j \sin(2j\pi t) \quad (2)$$

where a_j and b_j are the Fourier series parameters.

The seasonal variability is relevant for trends in atmospheric gases, and previous studies have shown that this variability is well captured by using the Fourier series of order 3 ($S=3$) (Gardiner et al., 2008; Franco et al., 2016; Prignon et al., 2019; Yamanouchi et al., 2021; Friedrich et al., 2020; Wizenberg et al., 2024). For this study, we apply Fourier series fitting of $S=3$ to the daily means of the NH_3 total column time-series data from the FTIR sites, GCHP, and TCR-2, to sites with more than 150 data points to ensure a good fit of the linear trend model. In addition, to estimate the uncertainty, we used bootstrap resampling methods, with a bootstrap population of 5000 as recommended in Gardiner et al. (2008), and an autoregressive wild bootstrap method (AWB) as recommended by Friedrich et al. (2020) to obtain the corresponding 95% confidence intervals (CI). The latter is more suitable for analyzing geophysical time series as it is specifically designed to account for missing observations,



auto-correlation, and seasonal effects in the datasets. Trends for which the CI does not include zero are considered statistically significant.

3 FTIR Results

3.1 Time Series

185 The retrieved total column time series of NH_3 from the 22 FTIR sites are shown in Figure 1. The NH_3 time series are very diverse across all sites, with Thule, Zugspitze, Toronto, Los Angeles, Izana, and Lauder, the only sites with more than 20 years of data. Most of the sites have between ten and 20 years of data, and only five sites have less than three years of data. The largest total columns of NH_3 are seen in Hefei, Bremen, Toronto, Mexico City and Porto Velho. These sites are in urban areas influenced by nearby agricultural activities, local sources, and biomass burning events. In contrast, the smallest total
190 columns of NH_3 are mainly at the Arctic sites, where the NH_3 sources are limited, and in Reunion Maido and Jungfraujoch, two high-altitude sites that most likely were measuring NH_3 background concentrations.

However, noticeable enhancements can be observed at some of the sites, such as Eureka, Thule, Izana, Los Angeles, Mexico City, Altzomoni, Paramaribo, Porto Velho and Reunion Maido, mainly attributed to biomass burning events. The NH_3 enhancements at Eureka and Thule were linked to the 2017 Canadian wildfires by Lutsch et al. (2019). A large fire that occurred in
195 the Amazon that reached several regions, analyzed by Bencherif et al. (2020), potentially reached Porto Velho and Paramaribo in August 2019. The NH_3 enhancements in late 2019 at the Reunion Maido site could be from biomass burning plumes from Africa and even the Amazon region (Callewaert et al., 2022).

Simultaneous enhancements of NH_3 and CO at Porto Velho and Reunion Maido during 2019 are shown in Figures A1 and A2 in the Appendix, as CO is largely emitted by fires: it has a longer lifetime than NH_3 and can be used as a reference species
200 (Whitburn et al., 2017). For Mexico City, the simultaneous enhancements of NH_3 and CO are shown in Figure A4 during May 2019; this enhancement was part of a pollution event further discussed in Rios et al. (2023). Unfortunately, there are no simultaneous measurements of CO at Altzomoni during the enhancement observed in late March 2019; however, a few fires in the surrounding areas were detected by the Aqua-MODIS instrument (<https://wvs.earthdata.nasa.gov>). Similarly, for the Los Angeles site, NH_3 and CO enhancements are observed during 2020 and 2021 (Figure A3, which can be attributed to the
205 exceptional California fires (Keeley and Syphard, 2021; Safford et al., 2022; Ayars et al., 2023).

For the Izana site, the enhancements observed in 2012 are from the Tenerife Island fire that reached the site in July 2012 (Santamarta Cerezal, 2013). In addition, in September 2021, a volcanic eruption on the island of La Palma (Canary Islands) affected the Izana site (García et al., 2023; Campeny et al., 2023). Studies on NH_3 emissions from volcanic eruptions are limited, but simultaneous enhancements of NH_3 and SO_2 from volcanoes has been reported (Uematsu et al., 2004; Adams
210 et al., 2017), Figure A5 shows SO_2 and NH_3 enhancements during the Tajogaite eruption from 19 September 2021 to 13 December 2021 during which volcanic plume reached the Izana site.

For the analysis in the following sections, the sites are divided into three categories: Arctic (Eureka, Ny Alesund, Thule), Northern Hemisphere (St. Petersburg, Bremen, Garmisch, Zugspitze, Jungfraujoch, Toronto, Boulder, Tsukuba, Los Angeles,

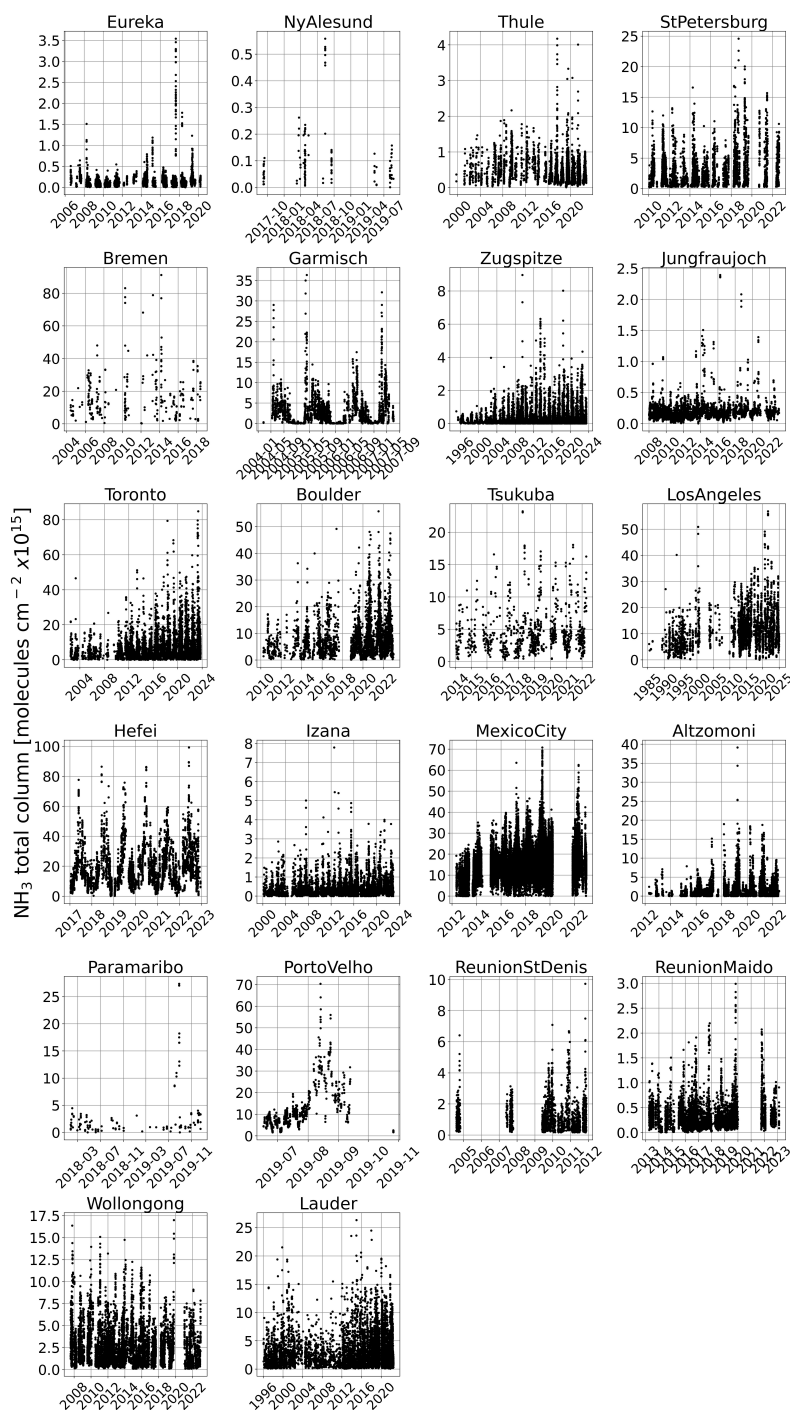


Figure 1. Time series of NH₃ total columns measured at the 22 FTIR sites.



Hefei, Izana, Mexico City, Alzomoni and Paramaribo), and Southern Hemisphere (Porto Velho, Reunion St. Denis, Reunion
215 Maido, Wollongong, and Lauder).

3.2 Seasonal Variability

The seasonal variability in NH_3 total columns corresponding to the monthly means calculated across all years is shown in
Figure 2. The seasonal patterns present similarities across most sites, with a few exceptions. In addition, the monthly means
are seen to increase for the most recent years for most sites. The monthly mean total columns vary between 0.03 and $42.38 \times$
220 10^{15} molec. cm^{-2} for all sites, with the maximum during spring and summer and the minimum in winter. For the Arctic sites,
the variability of NH_3 across the year (between February and October due to the lack of sunlight for FTIR measurements during
the Arctic winter) is small. The columns are between 0.06 and 0.70×10^{15} molec. cm^{-2} with the minimum in March and the
maximum in August at all three sites, and an average of 0.40×10^{15} molec. cm^{-2} , mainly due to NH_3 being transported from
225 large biomass burning events in northern latitudes, such as the August wildfires in 2017 (Lutsch et al., 2019). The seabirds'
nesting season in the Arctic occurs between May and September, and can be a significant N-input source of NH_3 to terrestrial
Arctic ecosystems (Wentworth et al., 2016). According to Riddick et al. (2012), the seabird colonies near Thule are larger than
those near Eureka, and the difference in the magnitude of the NH_3 columns between these sites is attributed to this source, as
shown by Lutsch et al. (2019), mainly due to the presence of seabird colonies on the Greenland coast (Murphy et al., 2025).

For the sites located in the Northern Hemisphere, excluding the Arctic sites, the monthly mean NH_3 total columns vary
230 between 0.03 and 42.38×10^{15} molec. cm^{-2} , with the largest values observed between April and August and the smallest values
during December and January. For most sites, NH_3 has a maximum in April and May, then decreases, with another maximum
in August in a few cases. However, the maximum monthly mean was found at Hefei in June (42.38×10^{15} molec. cm^{-2}) and
the minimum at Zugspitze in December (0.03×10^{15} molec. cm^{-2}). The observed pattern could reflect the influence of the
meteorological conditions, mainly related to the NH_3 temperature sensitivity, as warmer temperatures favour the conditions for
235 NH_3 volatilization (Sutton et al., 2013), and the rainfall, which favours wet deposition (Behera et al., 2013). Figure B3 shows
the monthly means of surface temperature derived from GCHP.

However, at some sites such as Boulder, a maximum was observed later in the year, probably due to the fertilizer application
season and agricultural practices. For Toronto, Mexico City, and Alzomoni, the seasonal patterns were directly compared with
those reported in Yamanouchi et al. (2021) and Herrera et al. (2022) as those studies also used these FTIR datasets. For Toronto,
240 the previously reported means were calculated between 2002 and 2018, while this study extends the time series up to 2023;
the patterns are very similar with the maximum in May and the minimum in January, however, the columns are larger for April
and June in the present study. For Mexico City and Alzomoni, the previously reported time series ended in 2020, while this
study extends the time series up to 2022; the patterns are similar with a small increase in the total column means in this study.

For the sites in the Southern Hemisphere, a similar pattern is seen, with more NH_3 during the warmest months, in this case
245 during the last months of the year, with overall monthly mean total columns between 0.21 and 27.82×10^{15} molec. cm^{-2} . The
maximum values were found at Porto Velho in August (27.82×10^{15} molec. cm^{-2}) and the minimum at Reunion Maido during
March (0.21×10^{15} molec. cm^{-2}). For Porto Velho, it is difficult to isolate a seasonal pattern due to the limited observations,



however, it is clear that August is the month with more NH_3 , potentially influenced by Amazonian biomass burning events, in agreement with a study by Whitburn et al. (2015), and specifically for 2019 when an exceptional wildfire event took place in the southern portion of the Amazon during August (Bencherif et al., 2020). Plumes from this fire event could have reached Paramaribo, explaining the enhancements in this site. Overall, the reported seasonal patterns are in agreement with other studies such as Van Damme et al. (2015), which used satellite data. However, this study provides details for the specific sites.

In addition, in Figure 4, the seasonal variability is also shown using a fitted Fourier series of third degree. It can be observed that Eureka, Jungfraujoch, and Reunion Maito have a small seasonal amplitude ($< 0.3 \times 10^{15}$ molec. cm^{-2}). Thule, Zugspitze, Izana, Alzomoni, Wollongong, and Lauder present a moderate seasonal amplitude (between 0.3 and 2×10^{15} molec. cm^{-2}). St. Petersburg, Garmisch, and Tsukuba show a large seasonal amplitude (between 2 and 6×10^{15} molec. cm^{-2}) and Bremen, Toronto, Boulder, Hefei, Los Angeles, and Mexico City have the largest seasonal amplitudes ($> 6 \times 10^{15}$ molec. cm^{-2}). Seasonal variability is markedly smaller at remote and high-altitude sites than at urbanized sites, highlighting the contrast between background atmospheric conditions and regions influenced by anthropogenic and biomass burning emissions.

3.3 Diurnal Variability

One advantage of the FTIR measurements, compared to satellite data, is their frequency, which allows diurnal cycles, during the sunlit part of the day, to be derived by calculating the hourly means as shown in Figure 3 in local hours for the 22 sites. The time series were deseasonalized (Varotsos et al., 2005; Vigouroux et al., 2015) before calculating the diurnal means by subtracting the seasonal component obtained through the evaluation of the fitted Fourier coefficients from Equation 2 at the timestamps of the FTIR time series to estimate the seasonal component at sub-daily resolution. The deseasonalized time series represents anomalies relative to the seasonal mean and can contain both positive and negative values. For Ny Alesund, Paramaribo, Porto Velho, and Reunion St. Denis, the fitting of Fourier series was not possible due to the limited amount of data; therefore, no deseasonalized time series were derived for these sites.

In this case, similar to the monthly means, larger columns are observed for the most recent years for most sites, however, the diurnal variability is very different across the sites, with hourly means between 0.004 and 30.33×10^{15} molec. cm^{-2} . Of the three Arctic sites, Thule showed the most complete diurnal cycle, while very little diurnal fluctuation is observed at these sites, with a mean of 0.47×10^{15} molec. cm^{-2} and a variation of less than 0.5×10^{15} molec. cm^{-2} during the day. This pattern is expected as there are limited sources in the Arctic region that change daily NH_3 concentrations. For the other sites located in the Northern Hemisphere, the diurnal means were between 0.004 and 30.33×10^{15} molec. cm^{-2} with the larger values in the afternoon between around 15h and the minimum in the early morning.

For the Southern Hemisphere, the diurnal means were between 0.10 and 13.68×10^{15} molec. cm^{-2} , smaller values than in the Northern Hemisphere, with no consistent patterns across the sites. However, Lauder presents a pattern similar to Alzomoni, with columns increasing steadily during the day until reaching a peak in the late afternoon. The variability of Alzomoni was explored in Herrera et al. (2022), where this pattern was attributed to NH_3 transported from surrounding urban areas. The high-altitude sites also present very little variability throughout the day, except for Alzomoni.

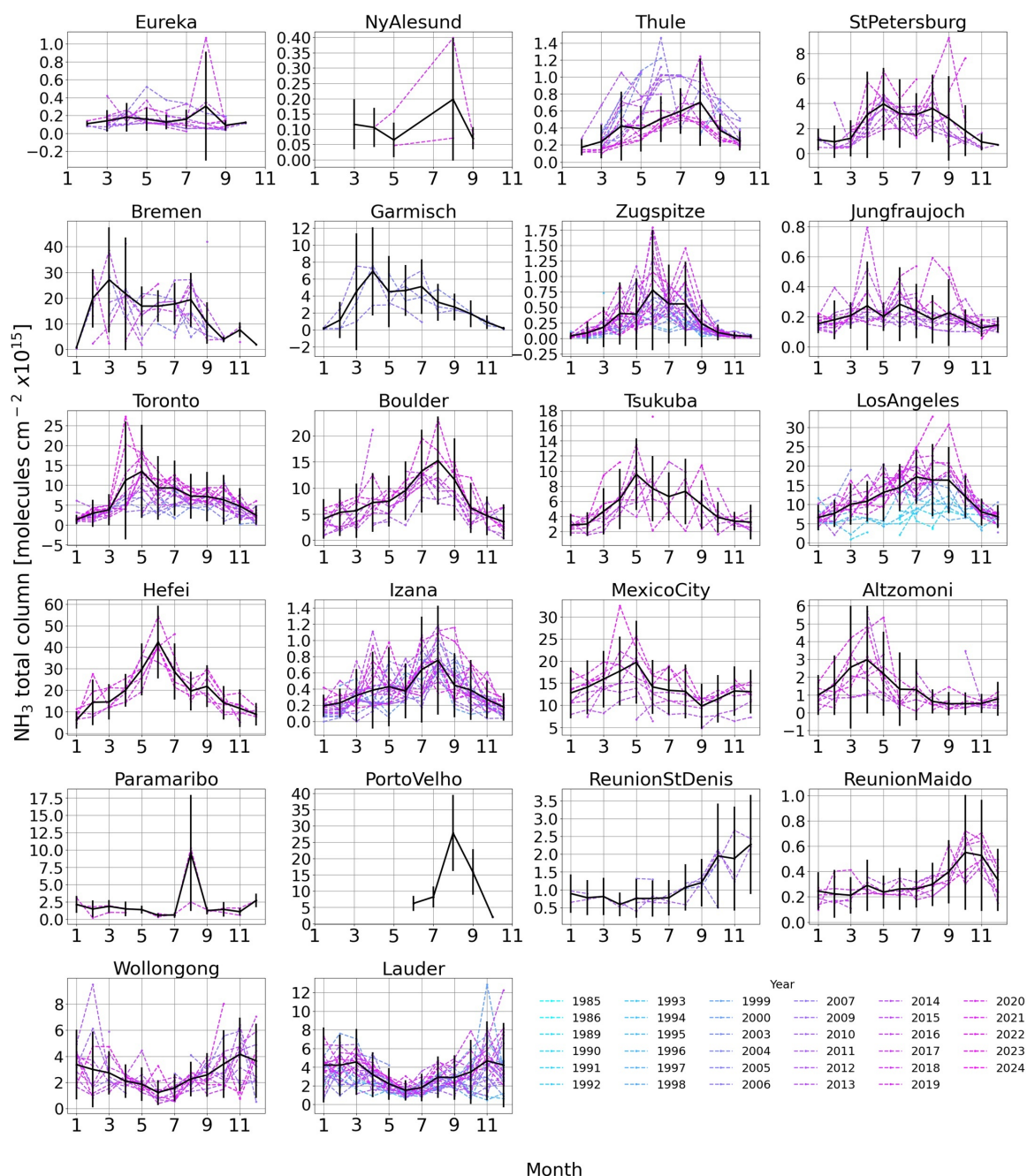


Figure 2. Monthly mean NH_3 total columns for each FTIR site. Different years are shown in different colours, with the most recent years in pink shades and older years in blue shades. The thick black line with error bars is the mean for all years $\pm 1\sigma$.



The reasons for the observed patterns vary depending on each site. Diurnal variations in the NH_3 total columns can be associated with vertical variations of the relative humidity and temperature profiles (Kutzner et al., 2021), as changes in these conditions as well as in the pH can lead to phase changes between NH_3 in the gas and particulate phase (Pye et al., 2020). In addition, transport from the surrounding regions plays a major role in the variability for some sites. Finally, for the sites located
285 in urban areas, traffic emissions during rush hour have an impact on the diurnal variability of NH_3 (Kotnala et al., 2019; Osada et al., 2019; Herrera et al., 2022; Gu et al., 2022; Viatte et al., 2023).

3.4 FTIR Trends

Long-term trends in the daily mean NH_3 total columns are calculated using Eqs. 1 and 2. The FTIR trends, the period used for the trend estimation, and confidence intervals for all sites are summarized in Table 2 and plotted in Figure 4. The mean trend
290 for all sites is $19.43 (14.18 - 24.68) \times 10^{13} \text{ molec. cm}^{-2} \text{ yr}^{-1}$ driven by the large trends at Hefei and Mexico City. Most of the sites present positive and significant trends. Only two sites present negative trends, Thule and Wollongong. Most of the trends are in agreement with trends reported by Van Damme et al. (2021) using ten years of satellite data. The drivers behind these trends vary between sites, and can be due to a change in NH_3 emissions, a change in meteorological conditions, or a change in the lifetime of NH_3 due to changes in NO_x and SO_2 abundances. For the Arctic sites, Thule presents the largest and most
295 significant trend, although it is negative. As mentioned in Section 3.2, seabird colonies are an important source of NH_3 at this site, so changes in the seabird population could impact the trend, such as the reported global decline in seabird population of 69.7% between 1950 and 2010 (Palczyński et al., 2015).

Europe is one of the few regions where actions towards the reduction of NH_3 emissions have been implemented. Zugspitze is located in Germany, where, according to the European Environment Agency (EAP) National Emission Reduction Commitments Directive reporting status 2022 (European Environment Agency, 2023), there have been reductions in NH_3 , NO_x , and
300 SO_2 emissions. In addition, Tichý et al. (2023) reported decreasing trends of NH_3 emissions over Europe of -26% over Europe using remote sensing data and inverse modelling analysis. However, the FTIR time series at the five European sites don't show this reduction, suggesting that more efforts focused on reducing NH_3 from the agricultural sector are needed.

Trends in reduced N have been previously reported in North America (Yao and Zhang, 2016; Yamanouchi et al., 2021; Benish et al., 2022; Herrera et al., 2022). In Canada, significant reductions in emissions of SO_x (79%) and NO_x (41%) between 1990
305 and 2021 have been reported (Government of Canada, 2023). In the US, the Acid Rain Program requires major emissions reductions in SO_2 and NO_x and has delivered annual SO_2 reductions of over 95% and NO_x reductions of over 89% (U.S. Environmental Protection Agency, 2024). These changes could be increasing the atmospheric lifetime of NH_3 over Toronto, Boulder, and Los Angeles, resulting in the positive NH_3 trends. Mexico City has the largest positive and significant trend; this
310 may be due to Mexican policies to regulate NO_x and SO_2 that (similar to Boulder and Toronto) could be affecting the lifetime of NH_3 .

For the Southern Hemisphere, the largest trend is from Wollongong, and it is negative. There have been recent efforts in Australia to improve agricultural practices to mitigate emissions of different gases, based on the implementation of technology and a best management practices approach (Panchasara et al., 2021). For NH_3 , the focus is on reducing the emissions from

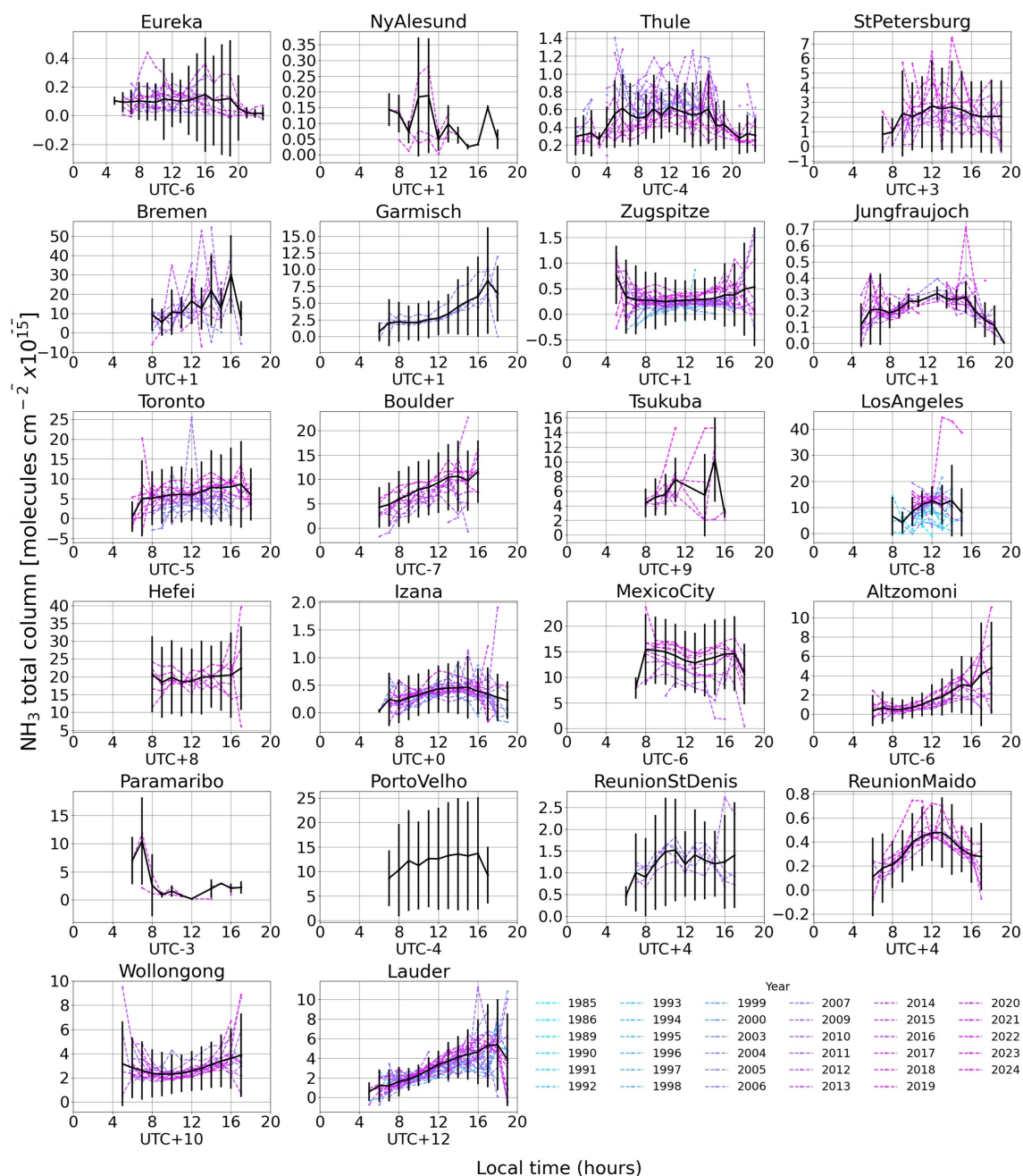


Figure 3. Hourly mean of deseasonalized NH_3 total columns in local time (hours, with offset from UTC for each site indicated on each panel) for each FTIR site. Different years are shown in different colours with the most recent years in pink shades and older years in blue shades. The thick black line with error bars is the mean for all years $\pm 1\sigma$.



Table 2. NH₃ mean total columns ± 1σ and absolute trends with 95% confidence intervals for the FTIR sites. Bold indicates a significant trend.

Station	Measurement	NH ₃ Mean Total Column (molec. cm ⁻²)×10 ¹⁵	NH ₃ Trend and 95% CI	
	Period		(molec. cm ⁻² yr ⁻¹)×10 ¹³	%
Eureka	2006 – 2020	0.17 ± 0.24	0.58 (0.13 – 1.02)	3.0 (0.73 – 5.21)
Ny Alesund	2017 – 2019	0.12 ± 0.12	–	–
Thule	1999 – 2022	0.46 ± 0.34	-2.10 (-2.50 – -1.70)	-7.7 (-9.1 – -6.25)
St. Petersburg	2010 – 2022	3.07 ± 2.85	11.70 (3.54 – 19.60)	4.67 (1.44 – 7.81)
Bremen	2004 – 2018	16.80 ± 14.70	28.20 (-8.20 – 63.80)	3.55 (-1.03 – 8.02)
Garmisch	2004 – 2007	3.56 ± 3.97	31.10 (-13.80 – 76.20)	10.64 (-6.39 – 28.04)
Zugspitze	1995 – 2023	0.32 ± 0.55	0.84 (0.58 – 1.10)	2.13 (1.43 – 2.84)
Jungfrauoch	2008 – 2023	0.20 ± 0.16	0.38 (0.13 – 0.63)	1.89 (0.66 – 3.07)
Toronto	2002 – 2023	7.98 ± 8.53	28.40 (21.20 – 35.40)	4.68 (3.53 – 5.82)
Boulder	2010 – 2022	9.00 ± 7.42	37.30 (26.30 – 48.30)	5.93 (4.11 – 7.75)
Tsukuba	2014 – 2022	4.64 ± 3.45	19.10 (7.02 – 31.40)	5.16 (1.82 – 8.46)
Los Angeles	1985 – 2024	11.31 ± 7.07	23.4 (19.60 – 27.40)	4.86 (4.07 – 5.69)
Hefei	2017 – 2022	19.20 ± 13.50	73.80 (16.60 – 132)	4.36 (0.78 – 7.90)
Izana	2000 – 2022	0.42 ± 0.47	0.38 (0.03 – 0.74)	0.88 (0.07 – 1.69)
Mexico City	2012 – 2022	14.50 ± 6.89	93.0 (78.8 – 107)	12.8 (10.86 – 14.79)
Altzomoni	2012 – 2022	1.63 ± 2.36	7.08 (2.56 – 11.50)	10.73 (3.94 – 17.46)
Paramaribo	2018 – 2019	3.02 ± 4.85	–	–
Porto Velho	2019	11.80 ± 9.82	–	–
Reunion St. Denis	2004 – 2011	1.21 ± 1.04	–	–
Reunion Maidu	2013 – 2023	0.34 ± 0.29	0.16 (-0.53 – 0.84)	0.49 (-1.73 – 2.59)
Wollongong	2007 – 2022	2.62 ± 2.12	-3.78 (-7.10 – -0.35)	-0.93 (-1.79 – -0.08)
Lauder	1996 – 2022	3.15 ± 2.95	2.37 (0.86 – 3.85)	1.63 (0.60 – 2.61)
Mean for all sites	–	–	19.43 (14.18 – 24.68)	3.82 (3.29 – 4.35)



315 volatilization by improving the irrigation methods, use of urease inhibitors, and use of ammonium-based fertilizers instead of
urea (Pan et al., 2016). Zhang et al. (2022) emphasized the costs and benefits of NH₃ abatement in this region because the
mitigation potential of NH₃ emissions from agricultural sources is around 32%. Similarly to Wollongong, several approaches
to mitigate NH₃ emissions have been analyzed for New Zealand (Jarvis and Ledgard, 2002; Parfitt et al., 2008; Saggar et al.,
2013). Despite this, the trend for Lauder is positive and significant. Around 50% of Australia's and New Zealand's land area
320 is used for agriculture (Pannell and Rogers, 2022). However, there are substantial differences in their agricultural sectors and
diversity, with more diversification in Australia and dominance of livestock grazing and dairy production in New Zealand
(Pannell and Rogers, 2022). Dairy farms involve several emission sources, such as grazing, fertilizer application, housed
animals, and stored and applied excreta (Jarvis and Ledgard, 2002). Implementing strategies to mitigate emissions from several
sources is more challenging, and this could be related to Lauder's positive trend. A more detailed discussion of similarities and
325 differences in policy approaches regarding the agricultural situation in New Zealand and Australia is discussed in Pannell and
Rogers (2022).

4 Comparison of Simulated NH₃ Total Columns

The FTIR measurements were used to evaluate the simulations of NH₃ by GCHP and TCR-2. Long-term time series of NH₃
total columns were extracted from the previously described GCHP model and TCR-2 product at the grid point closest to
330 the latitude and longitude of each FTIR station. The total columns from GCHP and TCR-2 were smoothed using the FTIR
averaging kernels (AVKs) and a priori profiles for each site following the methodology of Rodgers and Connor (2003). The
temporal coincidence criterion for matching the simulation and the FTIR observations was 2 h. The time series of NH₃ total
columns from FTIR, GCHP and TCR-2 are shown in Figure B2 of the Appendix.

To compare the simulated and FTIR total columns, correlations for the simulation-measurement pairs were performed, in
335 addition to the calculation of simulation minus measurement relative differences (RD) as defined by Equation 3 (Dammers
et al., 2016), where $TC_{S,i}$ and $TC_{F,i}$ are the total columns for the simulations and FTIR, respectively. The mean is calculated
to obtain the mean relative difference (MRD) for site i between the simulation and the FTIR measurements:

$$MRD = \frac{1}{N} \sum_{i=1}^N RD = \frac{1}{N} \sum_{i=1}^N \left(\frac{TC_{S,i} - TC_{F,i}}{TC_{F,i}} \right) \times 100. \quad (3)$$

The normalized root mean square error (NRMSE) was calculated with Equation 4, where N is the total number of model-
340 measurement pairs, and σ_F is the standard deviation of the FTIR data, following Kärnä and Baptista (2016) and Flood et al.
(2024):

$$NRMSE = \frac{1}{\sigma_F} \sqrt{\sum_{i=1}^N (TC_{S,i} - TC_{F,i})^2}. \quad (4)$$

A NRMSE close to 1 indicates a RMSE comparable to σ_F , $NRMSE > 1$ indicates a $RMSE > \sigma_F$ and poor fit, and $NRMSE$
345 < 1 indicates a $RMSE < \sigma_F$ and a better agreement of the simulation with respect to the FTIR. Linear regressions were fitted

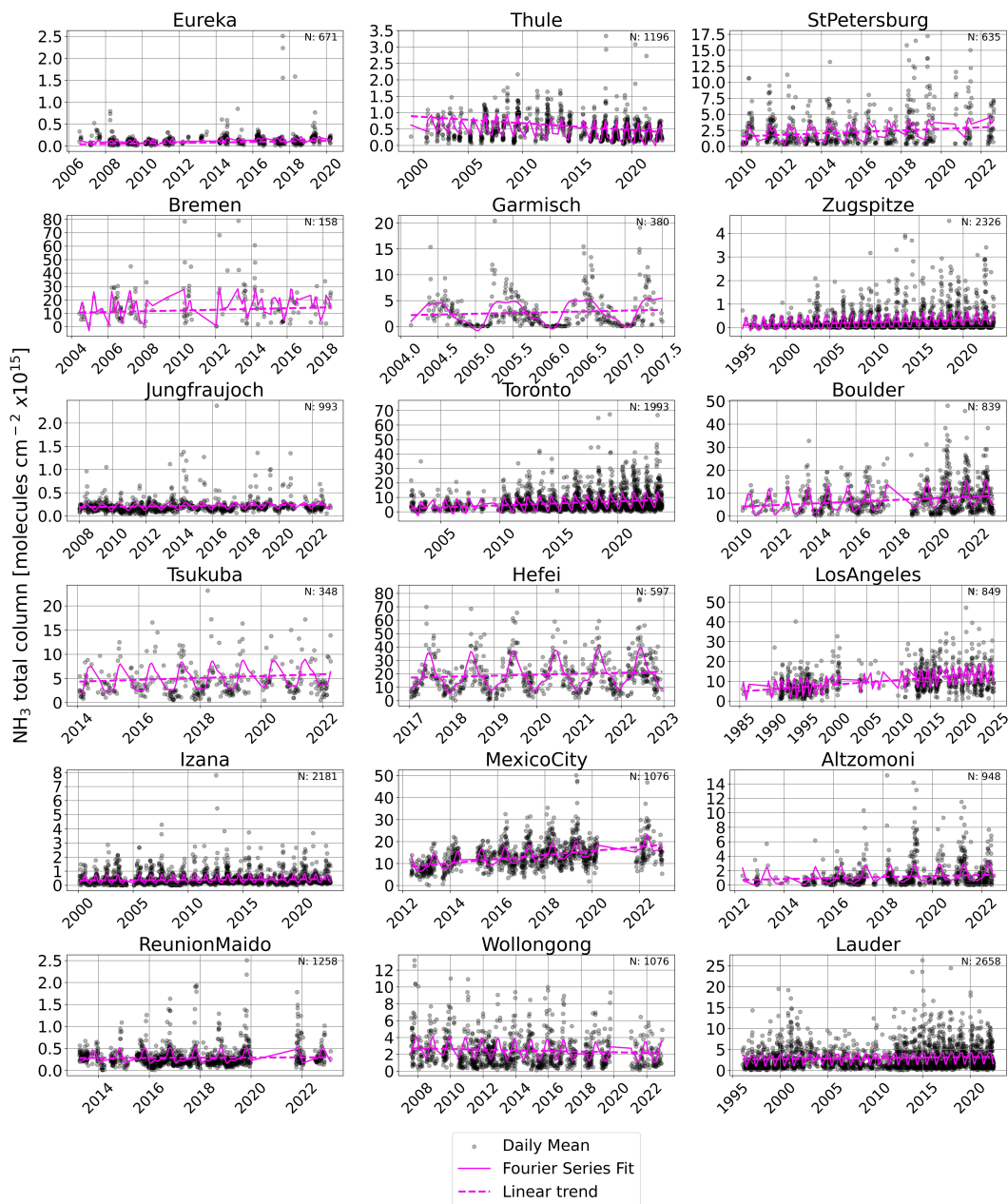


Figure 4. Time series of FTIR daily mean NH_3 total columns with fitted third-order Fourier series (solid blue line) and linear trends (dashed blue line) for the sites with sufficient data density (N) to derive trends.



to the scatter plot data of the GCHP and TCR-2 outputs versus the FTIR measurements, without forcing the intercept to zero. The results are summarized in Table 3, including the correlation coefficient R , the fitted slope, the MRD, and the NRMSE. Figure 5 shows the correlations between both datasets and the FTIR data at each station.

Table 3. Results of the NH_3 total column comparisons of GCHP and TCR-2 with the FTIR measurements.

Station	GCHP				TCR-2			
	R	Slope (molec. cm^{-2}) $\times 10^{15}$	MRD %	NRMSE	R	Slope (molec. cm^{-2}) $\times 10^{15}$	MRD %	NRMSE
Eureka	0.02	0.03	-30.18	1.97	0.81	0.05	-97.23	1.14
Ny Alesund	0.07	0.12	105.93	2.13	0.85	0.06	-73.63	1.19
Thule	0.09	0.47	316.31	6.60	0.12	0.02	-97.63	1.59
St. Petersburg	0.51	0.45	-18.63	0.96	0.39	0.16	-67.73	1.19
Bremen	0.12	0.07	-24.40	1.14	0.23	0.11	-14.45	1.02
Garmisch	0.28	0.50	217.42	2.64	0.18	0.20	111.83	1.66
Zugspitze	0.34	0.55	137.42	1.75	0.36	0.32	-0.32	1.07
Jungfraujoeh	0.32	0.34	-19.19	1.23	0.26	0.08	-91.97	1.46
Toronto	0.65	0.39	-44.90	0.88	0.36	0.22	-16.74	0.98
Boulder	0.50	0.34	-18.96	0.92	0.46	0.08	-88.03	1.40
Tsukuba	0.48	1.16	-5.65	2.11	0.45	0.47	-23.64	1.12
Los Angeles	0.55	0.24	-64.47	1.37	0.45	0.26	-42.63	1.20
Hefei	0.40	0.86	-18.78	1.96	0.07	0.15	97.18	2.56
Izana	0.18	0.19	-16.12	0.61	-0.04	0.11	-96.18	0.65
Mexico City	0.38	0.13	-79.02	1.89	0.16	0.03	-88.09	2.13
Altzomoni	0.49	0.11	-73.93	1.04	0.22	0.00	-96.01	1.21
Paramaribo	0.08	0.01	-85.00	1.13	0.09	0.00	-97.06	1.72
Porto Velho	0.85	0.68	-31.02	0.64	NA	NA	NA	NA
Reunion St. Denis	0.22	0.05	-80.41	1.34	-0.02	0.00	-74.63	1.33
Reunion Madio	0.59	0.11	-83.87	1.33	0.13	0.00	-98.86	1.60
Wollongong	0.49	0.37	-11.36	0.91	0.23	0.09	-62.69	1.26
Lauder	0.28	0.12	-56.70	1.15	0.25	0.09	-55.49	1.13
Mean for all sites	0.36	0.27	0.66	1.62	0.28	0.22	-51.14	1.36

The comparisons vary from site to site and differ between GCHP and TCR-2. For GCHP, R varies from a minimum of 0.02 at Eureka, to a maximum of 0.85 at Porto Velho. For TCR-2, R varies from a minimum of -0.02 at Reunion St. Denis, to a maximum of 0.85 at Ny Alesund. GCHP overestimates NH_3 at two of the three Arctic sites, with poor correlations, large NRMSE, and large columns in the lower range of FTIR, while TCR-2 underestimates NH_3 at all three sites, but with better correlations and NRMSE. For the other Northern Hemisphere sites, the correlation coefficients for GCHP improved,



with larger $R = 0.65$ for Toronto. GCHP underestimates NH_3 for all sites except Garmisch and Zugspitze, where it significantly
355 overestimates the NH_3 column with large NRMSE values. For TCR-2, the correlations are mostly smaller than those for GCHP,
with negative MRDs, a maximum R of 0.46 for Boulder, and a minimum of -0.04 at Izana, while the highest NRMSE are for
Hefei and Mexico City. For the Southern Hemisphere, GCHP correlation coefficients are between $R = 0.85$ and 0.22, with the
best correlation for Porto Velho and the smallest value for Reunion St. Denis, with negative MRDs and NRMSE values close
to one. For TCR-2, similarly to the Northern Hemisphere sites, the correlations are small, between $R = 0.25$ for Lauder and R
360 = -0.020 for Reunion St. Denis. TCR-2 continues underestimating NH_3 with negative MRDs and $\text{NRMSE} > 1$.

Using all the available coincidences for all sites, there is an overall correlation coefficient of $R = 0.36$ and a slope of 0.27
(± 0.002) for GCHP and $R = 0.28$ and a slope of 0.22 (± 0.003) for TCR-2 as shown in Figure B1 of the Appendix. Figure 5
shows that the GCHP and TCR-2 total columns are larger than the FTIR measurements at the lower range of values, with larger
columns from TCR-2 than GCHP. Both model datasets performed similarly at the higher range of values, with lower columns
365 than the FTIR values.

4.1 Seasonal Variability

Figure 6 shows the FTIR, GCHP, and TCR-2 monthly mean NH_3 total columns, including the temporally matched FTIR and
models data (solid lines) and the complete time series (dashed lines) between 2003-2021 for GCHP and 2005-2018 for TCR-2.
The RD was calculated for each month for each site; then, using the monthly means, the MRD was obtained as reported in Table
370 4. Overall, no major differences were observed between the dashed and solid lines in Figure 6. Both GCHP and TCR-2 captured
similar variability as the FTIRs; however, there are some sites where the differences are more noticeable. Using data for all
sites, the analysis suggested a monthly MRD = 0.41% for GCHP and -49.25% for TCR-2. For the Arctic sites, GCHP mainly
overestimates and TCR-2 consistently underestimates NH_3 compared to the FTIR observations, while GCHP captures more
variability than TCR-2. For the rest of the Northern Hemisphere sites, the best agreements were for Jungfraujoch, Boulder, and
375 Tsukuba for GCHP, and Bremen, Zugspitze, Toronto, and Tsukuba for TCR-2, respectively, but overall, both datasets mainly
underestimate NH_3 at all sites. For the Southern Hemisphere, GCHP and TCR-2 presented similar results. For Porto Velho,
GCHP and TCR-2 captured the NH_3 maximum between August and September; moreover, GCHP was able to capture the
August 2019 enhancement. Both datasets underestimated NH_3 and barely captured the variability for Reunion St. Denis and
Reunion Maito. For Wollongong, GCHP is in good agreement with FTIR, while TCR-2 captures the variability but with an
380 underestimation, as in Lauder, where both datasets represented the variability but underestimated the monthly means.

4.2 Diurnal Variability

The NH_3 simulations do not have diurnal variation in anthropogenic emissions; however, in this section, comparisons of the
hourly means were performed to assess the ability of GCHP and TCR-2 to capture diurnal variability based on all the other
processes involved in the simulations, such as the chemistry, transport, and meteorology variations. For this section, the FTIR
385 and simulated time series of NH_3 total columns were not deseasonalized, as the deseasonalized and regular time series were

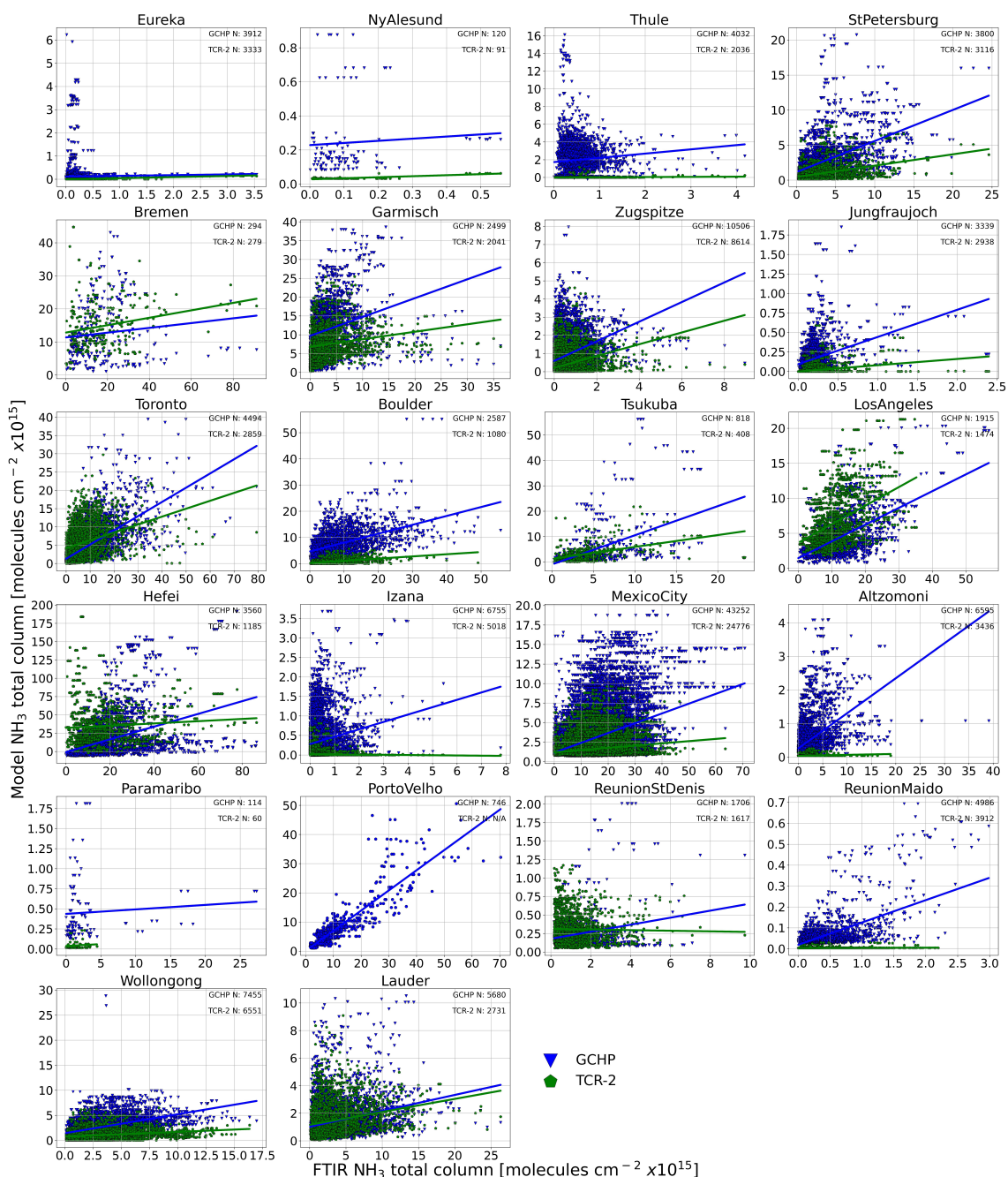


Figure 5. Scatter plots of GCHP and TCR-2 vs. FTIR NH_3 total columns. The lines show the individual regression results for each dataset. Note that for Porto Velho only the correlation between FTIR and GCHP is shown, as there are no TCR-2 data available for this site.



Table 4. MRD for monthly means of GCHP and TCR-2 NH₃ total columns compared to FTIR measurements at each site.

Station	GCHP MRD %	TCR-2 MRD %
Eureka	-41.87	-97.06
Ny Alesund	128.64	-77.99
Thule	265.41	-97.81
St. Petersburg	-23.21	-59.97
Bremen	-21.95	-5.84
Garmisch	233.47	132.21
Zugspitze	140.81	-2.19
Jungfrauoch	-9.88	-90.98
Toronto	-46.41	-18.35
Boulder	-18.12	-88.79
Tsukuba	1.46	-15.92
Los Angeles	-62.78	-42.66
Hefei	-22.92	93.02
Izana	-21.63	-87.18
Mexico City	-80.06	-87.98
Altzomoni	-72.85	-95.62
Paramaribo	-78.99	-96.05
Porto Velho	-28.12	NA
Reunion St. Denis	-80.81	-72.30
Reunion Mado	-84.11	-98.80
Wollongong	-11.69	-66.89
Lauder	-55.28	-57.11
All sites	0.41	-49.25

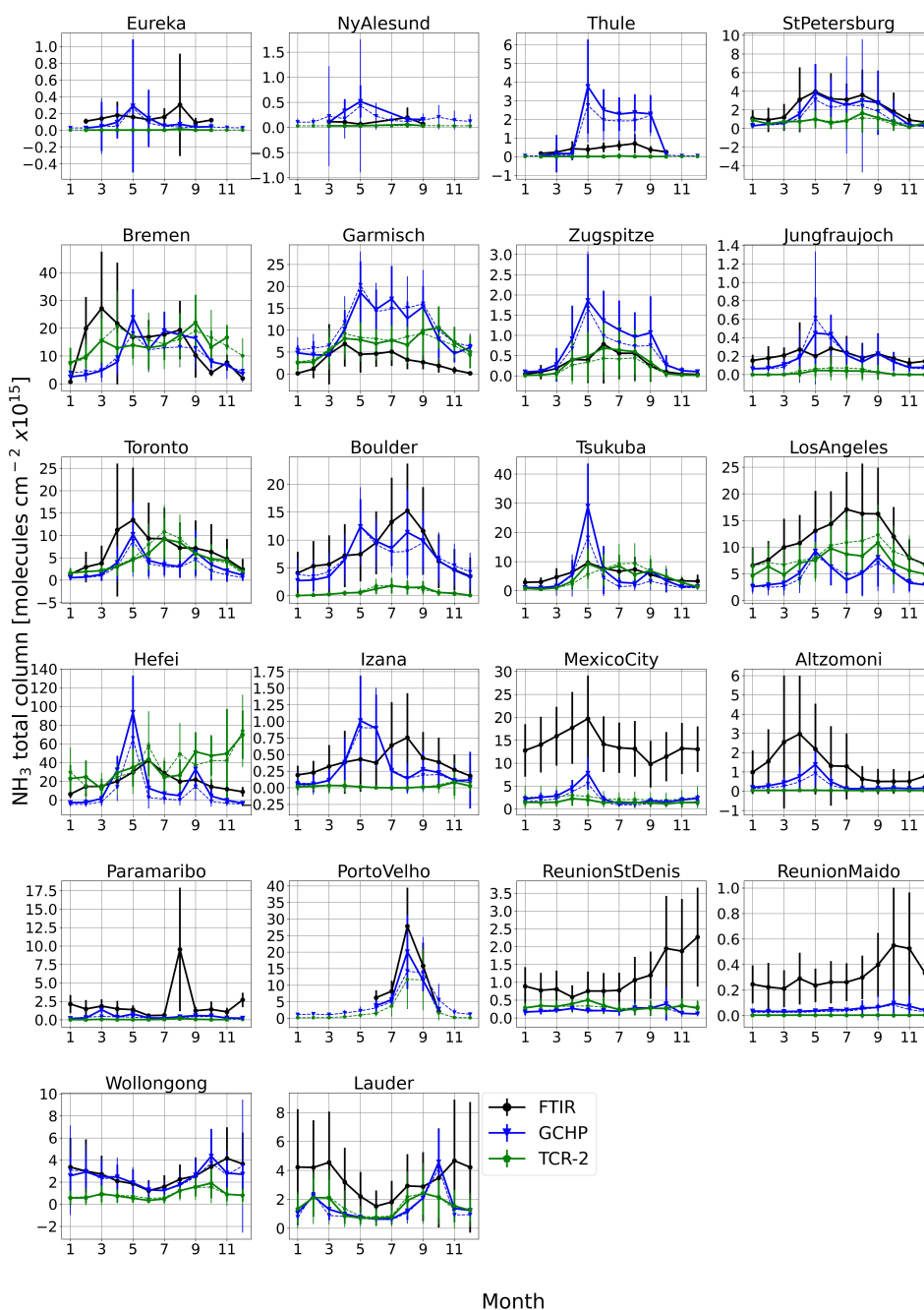


Figure 6. Monthly mean NH_3 total columns FTIR (black), GCHP (blue), and TCR-2 (green). The thick solid lines represent the temporally matched FTIR measurements and GCHP and TCR-2 datasets; the dashed lines represent the complete time series from each dataset. Vertical lines represent the standard deviation ($\pm 1\sigma$).



comparable with a mean $R = 0.92$ for all sites. The hourly means are plotted in Figure 7; the RD was calculated for the matching hours for each site, then the MRD of these hourly means was obtained as shown in Table 5.

There are large differences between the simulated and the measured total columns, as both GCHP and TCR-2 struggled to capture the diurnal variability of NH_3 , with an hourly MRD for all sites of 25.47% for GCHP and -47.84% for TCR-2. For the Arctic sites, TCR-2 doesn't capture any diurnal variability, while GCHP seems to capture some variability but with large overestimations, mainly at Ny Alesund and Thule. GCHP performed better at St. Petersburg and Wollongong and TCR-2 at Zugspitze where both datasets effectively reproduced the diurnal cycles, however, in most of the cases, there are large negative biases as observed in Figure 7 and Table 5, except for TCR-2 at Hefei and Garmisch where there is overestimation for the matching time series but better agreement with the full time series. The 24-hour variability from GCHP and TCR-2 could be relevant for some sites, but overall both simulations are unable to capture the diurnal variability of the FTIR instruments.

Table 5. MRD for diurnal means of GCHP and TCR-2 NH_3 total columns compared to FTIR measurements at each site.

Station	GCHP MRD %	TCR-2 MRD %
Eureka	19.91	-96.83
Ny Alesund	234.15	-46.67
Thule	494.09	-97.13
St. Petersburg	9.50	-58.29
Bremen	-35.23	21.35
Garmisch	261.96	109.41
Zugspitze	122.25	-2.38
Jungfraujoch	-36.98	-88.93
Toronto	-40.25	-23.19
Boulder	-16.39	-87.80
Tsukuba	70.75	-35.75
Los Angeles	-42.38	-42.61
Hefei	-22.83	92.72
Izana	-21.35	-96.05
Mexico City	-80.67	-84.95
Altzomoni	-73.51	-96.00
Paramaribo	-39.68	-94.66
Porto Velho	-26.96	NA
Reunion St. Denis	-79.91	-71.91
Reunion Maida	-83.98	-98.71
Wollongong	-9.89	-64.02
Lauder	-42.25	-42.21
All sites	25.47	-47.84

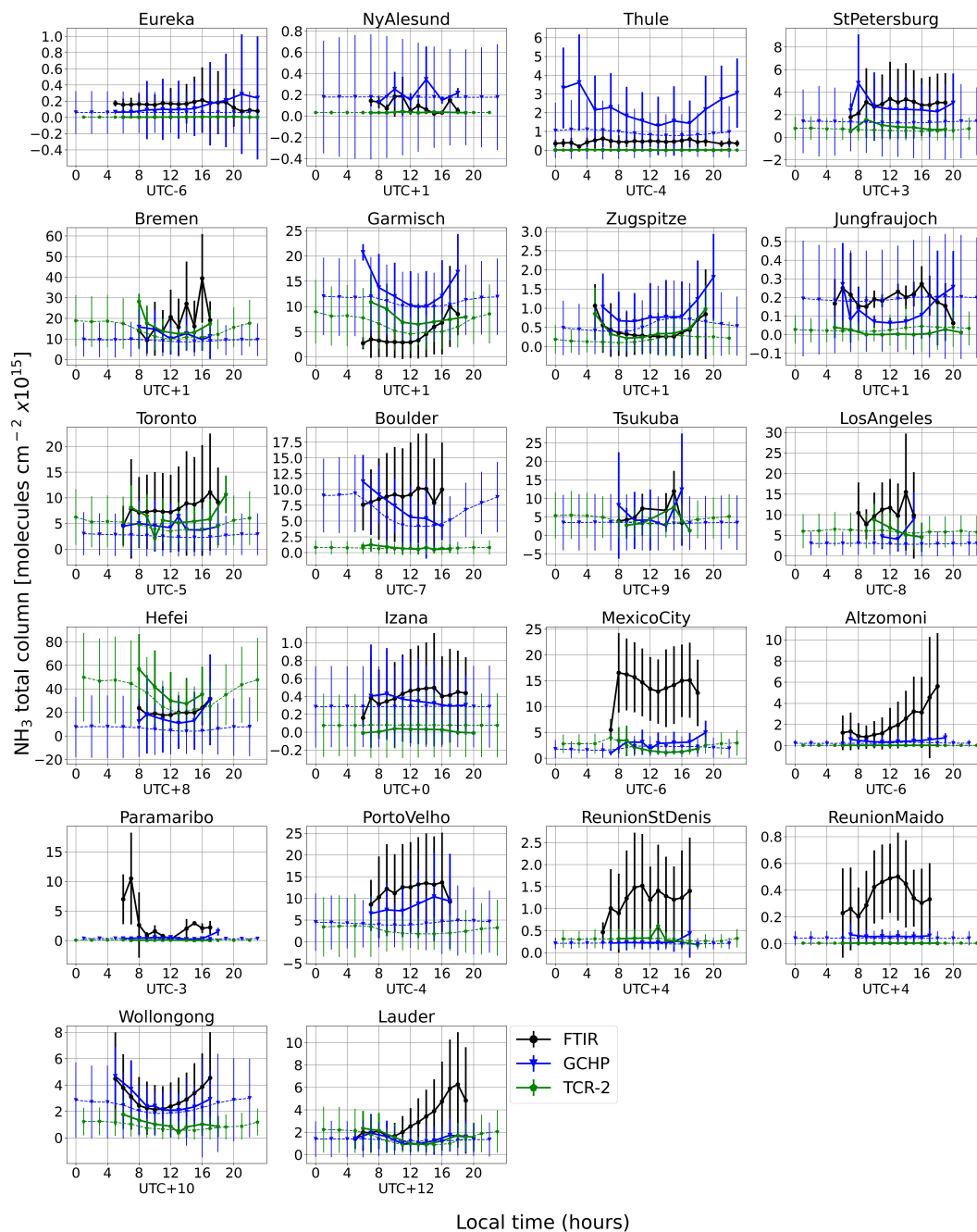


Figure 7. Hourly mean NH_3 total columns of FTIR (black), GCHP (blue), and TCR-2 (green) in local time (hours, with offset from UTC for each site indicated on each panel) for each site. The thick solid lines represent the temporally matched datasets between the FTIR measurements and GCHP and TCR-2 datasets; the dashed lines represent the complete time series from each dataset. Vertical lines represent the standard deviation ($\pm 1\sigma$).



4.3 Sensitivity Tests

Overall, GCHP NH₃ total columns agree better with the FTIR measurements than does TCR-2 for most sites, despite TCR-2 having higher spatial resolution and assimilated observations of relevant tropospheric species. Regardless of the optimization of the emissions and concentrations of various trace gases performed by TCR-2, NH₃ is not among the species that were
400 assimilated. Consequently, the emissions of the TCR-2 NH₃ product in this work are not optimized, and its concentration is only indirectly improved by optimizing species related to the NH₃ chemistry, such as NO_x and SO₂.

One difference between TCR-2 and GCHP is the a priori input emissions inventory used: CEDS for GCHP and HTAP for TCR-2 as described in Section 2.2. To explore the impact of the emissions inventory, we performed sensitivity tests for GCHP
405 by running the model using HTAP emissions for one month (February 2013). In addition, the stand-alone input emissions from HTAP and CEDS were analyzed independently. The MRD difference for the GCHP-HTAP vs. GCHP-CEDS NH₃ total columns and between HTAP and CEDS NH₃ total emissions are presented in Table 6 and were calculated using:

$$MRD = \frac{1}{N} \sum_{i=1}^N RD = \frac{1}{N} \sum_{i=1}^N \left(\frac{HTAP_{TC/Emissions,i} - CEDS_{TC/Emissions,i}}{CEDS_{TC/Emissions,i}} \right) \times 100. \quad (5)$$

The results show that the GCHP-HTAP NH₃ total columns were, on average 58.90% larger than the respective GCHP-CEDS columns for all sites. The sites with negative HTAP-CEDS MRD in the total columns were Eureka, Tsukuba, and all sites with
410 a latitude south of Izana, while Bremen, Garmisch, Zugspitze, Jungfrauoch, Toronto, Boulder and Los Angeles presented large positive MRD for the total columns.

The total HTAP NH₃ emissions are 10.85% larger than the total CEDS emissions for all sites, with the biggest differences for Los Angeles, Eureka, Hefei, Boulder and St. Petersburg. Among the emissions, the contribution from anthropogenic and seabird emissions the most different sectors.

This sensitivity test suggests that the larger underestimation in the NH₃ total columns from TCR-2 compared with GCHP is
415 not solely due to the a priori input emissions, since the total columns of GCHP-HTAP were, on average, larger than GCHP-CEDS despite the HTAP emissions being smaller than the CEDS emissions. The systematic negative bias in TCR-2 relative to the FTIR measurements can be related to persistent challenges common to many CTMs. Bian et al. (2017) highlighted that negative biases in NH₃ simulations can result from significant uncertainties in the wet deposition process, where the effective
420 Henry's Law constant for NH₃ varies significantly among different global models, up to six orders of magnitude, depending on what was considered in each model for its calculation (Bian et al., 2017). Additionally, the lack of NH₃ recycling between the atmosphere and soil further contributes to these biases. The implementation of a bi-directional flux scheme has been shown to mitigate these biases, as demonstrated in Zhu et al. (2015) and Cao et al. (2022) for the GEOS-Chem adjoint model. However, this scheme is not included in many CTMs, including the forecast model of TCR-2, limiting its ability to correct for such
425 biases. In the TCR-2 framework, positive model biases in sulfate and nitric acid could also lead to an underestimation of the ratio of gaseous ammonia to particulate ammonium. This issue is partially addressed by optimizing SO₂ and NO_x emissions through data assimilation (Miyazaki et al., 2020a; Sekiya et al., 2023). To further improve the representation of NH₃ in the data



assimilation framework, the most effective approach would be to incorporate improved model schemes while simultaneously assimilating SO₂ and NO₂, along with NH₃ observations.

Table 6. Mean relative differences between GCHP-HTAP and GCHP-CEDS calculated using Equation 5 for NH₃ total columns and NH₃ total emissions for February 2013.

Station	MRD columns %	MRD emissions %
Eureka	-12.01	-99.34
Ny Alesund	9.87	-0.93
Thule	9.00	0.60
St. Petersburg	21.81	-55.79
Bremen	195.69	49.56
Garmisch	210.17	20.45
Zugspitze	210.17	20.45
Jungfraujoch	175.00	27.16
Toronto	121.30	26.21
Boulder	220.52	81.03
Tsukuba	-12.66	-39.37
Los Angeles	213.70	267.64
Hefei	19.87	98.17
Izana	14.31	-67.71
Mexico City	-4.82	-4.11
Altzomoni	-4.82	-4.11
Paramaribo	-14.53	-27.20
Porto Velho	-14.93	-14.77
Reunion St. Denis	-19.20	-7.29
Reunion Maito	-19.20	-7.29
Wollongong	-13.19	-12.05
Lauder	-8.50	-12.64
All sites	58.98	10.85

430 4.4 Trend Comparison

Long-term trends were derived from the FTIR, GCHP, and TCR-2 datasets using temporally matched daily means as shown in Table 7. The FTIR trends using only the matching data do not indicate major changes compared with the trends in Table 2. The overall GCHP trend is $7.30 (-2.45 - 17.04) \times 10^{13}$ molec. cm⁻² yr⁻¹, smaller than the corresponding FTIR value, while the TCR-2 trend is $36.28 (-7.00 - 79.57) \times 10^{13}$ molec. cm⁻² yr⁻¹, larger than the FTIR trend, mainly due to the large TCR-2



435 trend at Hefei. For GCHP, the trend magnitudes are similar to the FTIR trends; however, there are changes in the statistical significance, with the largest differences for the Arctic and Southern Hemisphere sites. For TCR-2, most of the trends were positive but only two were significant. Both GCHP and TCR-2 have differences with the FTIR trends; however, the performance of GCHP is more consistent with the FTIR results than that of TCR-2.

Table 7. Trends in daily mean NH₃ total columns and 95% confidence intervals from GCHP and TCR-2 for daily means temporally matched to the FTIR measurements along with the corresponding FTIR trends. The model profiles were smoothed by the FTIR AVKs prior to calculating total columns. Bold indicates a significant trend.

Station	Time period	NH ₃ Trends and 95% CI (molec. cm ⁻² yr ⁻¹) × 10 ¹³				
		FTIR	GCHP	Time	FTIR	TCR-2
Eureka	2006-2020	0.58 (0.13 – 1.01)	1.12 (-0.31 – 2.58)	2006 -2018	0.48 (-0.10 – 1.07)	0.02 (-0.01 – 0.05)
Thule	2003-2021	-2.25 (-2.72 – -1.78)	-1.22 (-2.91 – 0.44)	2005-2018	-2.44 (-3.21 – -1.67)	0.06 (-0.01 – 0.14)
St. Petersburg	2010-2021	15.1 (5.52 – 24.3)	9.72 (1.34 – 18.4)	2010-2018	6.83 (-3.27 – 15.7)	-3.34 (-7.34 – 0.60)
Bremen	2004-2018	28.2 (-8.12 – 64.4)	45.1 (17.2 – 74.5)	2005-2018	18.4 (-19.6 – 58.1)	-6.88 (-30.5 – 16.5)
Garmisch	2004-2007	30.1 (-17.3 – 76.6)	62.2 (-17.9 – 145)	2005-2007	50.7 (-2.67 – 130)	56.4 (-8.59 – 122)
Zugspitze	2003-2021	0.82 (0.40 – 1.25)	3.30 (2.64 – 3.98)	2005-2018	1.32 (0.68 – 1.95)	0.18 (-0.34 – 0.70)
Jungfraujoch	2008-2021	0.52 (0.21 – 0.82)	1.61 (1.29 – 1.94)	2008-2018	0.49 (0.09 – 0.88)	0.09 (-0.05 – 0.23)
Toronto	2003-2021	31.7 (24.7 – 38.8)	17.5 (13.1 – 21.8)	2005-2018	30.6 (19.5 – 41.6)	4.20 (-1.54 – 9.86)
Boulder	2010-2021	45.3 (30.5 – 59.6)	21.7 (11.8 – 31.6)	2010-2018	30.6 (5.92 – 55.9)	1.07 (-3.27 – 5.42)
Tsukuba	2014-2021	16.1 (2.66 – 29.1)	12.6 (-14.5 – 40.6)	2014-2018	-6.09 (-33.3 – 21.9)	25.1 (3.45 – 45.8)
Los Angeles	2003-2021	20.9 (3.64 – 3.85)	16.0 (9.24 – 22.9)	2005-2018	34.0 (8.07 – 59.4)	9.09 (-6.18 – 24.30)
Hefei	2017-2021	67.8 (-2.02 – 135)	-74.8 (-213 – 64.7)	2017-2018	93.9 (-207 – 395)	564 (-154 – 1310)
Izana	2003-2021	0.32 (-0.17 – 0.82)	1.69 (1.30 – 2.10)	2005-2018	0.35 (-0.37 – 1.16)	-0.01 (-0.14 – 0.10)
Mexico City	2012-2021	107 (90.2 – 124)	6.70 (-0.02 – 13.7)	2012-2018	119 (100 – 137)	1.88 (-1.63 – 5.46)
Altzomoni	2012-2021	10.7 (5.80 – 15.6)	0.98 (-0.06 – 2.03)	2012-2018	-0.07 (-6.64 – 6.45)	0.05 (-0.02 – 0.12)
Reunion Maïdo	2013-2021	0.17 (-0.75 – 1.11)	0.26 (0.08 – 0.45)	2013-2018	-1.27 (-2.17 – -0.34)	0.005 (0.001 – 0.01)
Wollongong	2007-2021	-3.34 (-7.17 – 0.52)	4.17 (1.38 – 7.02)	2007-2018	-4.35 (-9.15 – 0.63)	0.92 (-0.93 – 2.80)
Lauder	2003-2021	2.07 (-0.26 – 4.42)	2.95 (1.75 – 4.16)	2005-2018	8.81 (4.96 – 12.6)	0.26 (-1.23 – 1.75)
Mean all	-	20.66 (15.02 – 26.30)	7.30 (-2.45 – 17.04)	-	21.18 (2.65 – 39.71)	36.28 (-7.00 – 79.57)

440 The NH₃ total column trends derived from the full-time series of GCHP and TCR-2 for all sites are shown in Table 8. Overall, most of the GCHP trends are positive and significant, while for TCR-2, there are more negative and fewer significant trends. In addition, GCHP trends are much larger in magnitude than those for TCR-2, and there are differences compared to the trends in Table 7. For GCHP, all the trends for the Arctic sites are small, positive and non-significant; for the Northern Hemisphere all trends were positive and significant, including Hefei; for the Southern Hemisphere only Porto Velho presented a negative trend that is non-significant, while the other sites presented positive trends, including Wollongong. For the Arctic sites, TCR-2 trends are very small and non-significant; for the Northern Hemisphere sites the trends are small and just a few



Table 8. GCHP and TCR-2 trends in NH₃ total columns derived from full time series of each model along with 95% confidence intervals. Bold indicates a significant trend. Model values correspond to the nearest model grid cell to each site (no interpolation applied).

Station	GCHP (2003–2021) NH ₃ trends and 95% CI		TCR-2 (2005–2018) NH ₃ trends and 95% CI	
	(molec. cm ⁻² yr ⁻¹)×10 ¹³	%	(molec. cm ⁻² yr ⁻¹)×10 ¹³	%
Eureka	0.87 (-0.42 – 2.19)	1.99 (-0.97 – 5.01)	0.03 (-0.01 – 0.07)	72.90 (-11.00 – 156.00)
Ny Alesund	0.90 (0.12 – 1.67)	3.42 (0.44 – 6.38)	0.05 (-0.01 – 0.11)	29.64 (-3.90 – 62.00)
Thule	0.81 (-0.24 – 1.91)	1.05 (-0.31 – 2.47)	0.03 (0.00 – 0.07)	31.11 (2.06 – 58.90)
St. Petersburg	3.02 (1.69 – 4.27)	4.09 (2.29 – 5.79)	-1.12 (-2.13 – -0.14)	-1.77 (-3.36 – -0.21)
Bremen	43.80 (39.60 – 48.30)	6.34 (5.73 – 6.98)	5.46 (0.98 – 9.99)	0.59 (0.11 – 1.08)
Garmisch	19.10 (16.80 – 21.70)	4.23 (3.72 – 4.79)	4.10 (0.97 – 7.16)	0.92 (0.22 – 1.60)
Zugspitze	19.10 (16.80 – 21.50)	4.23 (3.70 – 4.76)	3.58 (0.68 – 6.38)	0.90 (0.17 – 1.61)
Jungfraujoch	25.30 (21.80 – 28.60)	4.74 (4.10 – 5.36)	1.38 (-1.36 – 4.22)	0.37 (-0.36 – 1.12)
Toronto	12.20 (10.50 – 14.00)	10.14 (8.73 – 11.60)	2.58 (-0.01 – 5.12)	0.68 (0.00 – 1.35)
Boulder	12.50 (10.90 – 14.30)	6.40 (5.54 – 7.29)	0.30 (-0.30 – 0.91)	0.54 (-0.54 – 1.62)
Tsukuba	8.18 (5.50 – 11.00)	3.80 (2.55 – 5.10)	12.80 (9.49 – 16.10)	5.04 (3.74 – 6.34)
Los Angeles	7.75 (5.24 – 10.20)	2.66 (1.80 – 3.50)	-1.55 (-4.85 – 1.81)	-0.26 (-0.80 – 0.30)
Hefei	41.20 (27.70 – 54.40)	5.48 (3.69 – 7.24)	23.70 (9.56 – 37.20)	1.50 (0.61 – 2.36)
Izana	3.91 (2.99 – 4.81)	8.58 (6.57 – 10.60)	-0.19 (-0.51 – 0.12)	-1.59 (-4.38 – 1.05)
Mexico City	6.20 (4.41 – 7.98)	3.62 (2.57 – 4.66)	-1.77 (-2.37 – -1.18)	2.58 (-3.46 – -1.72)
Altzomoni	6.16 (4.32 – 7.95)	3.60 (2.52 – 4.64)	-1.77 (-2.37 – -1.15)	2.58 (-3.46 – -1.68)
Paramaribo	0.64 (0.32 – 0.95)	1.24 (0.62 – 1.84)	-0.13 (-0.21 – -0.05)	-2.10 (-3.42 – -0.81)
Porto Velho	-5.84 (-10.60 – -1.12)	-1.36 (-2.47 – -0.26)	-5.55 (-11.00 – 0.03)	-1.63 (-3.23 – 0.01)
Reunion St. Denis	0.11 (0.04 – 0.19)	0.67 (0.21 – 1.12)	1.09 (0.94 – 1.24)	6.51 (5.61 – 7.43)
Reunion Maito	0.11 (0.04 – 0.18)	0.65 (0.21 – 1.09)	1.09 (0.94 – 1.24)	6.51 (5.61 – 7.44)
Wollongong	2.69 (0.29 – 5.17)	1.60 (0.17 – 3.08)	-1.65 (-2.46 – -0.85)	-1.95 (-2.90 – -1.01)
Lauder	4.42 (3.31 – 5.54)	3.36 (2.52 – 4.22)	-0.02 (-0.74 – 0.68)	-0.02 (-0.59 – 0.54)
Mean for all sites	10.11 (9.31 – 10.91)	3.66 (3.35 – 3.97)	1.93 (1.11 – 2.75)	6.49 (2.00 – 10.98)



of them are significant, including the negative trend for Mexico City; finally for the Southern Hemisphere, the trend for Porto Velho is also negative but non-significant, while the trend for Wollongong is negative, significant and similar to the FTIR trend.

The NH_3 partitioning at each site was derived using GCHP total columns of NH_3 and NH_4^+ to calculate the $\text{NH}_3/\text{NH}_4^+$ ratio and explore which was the predominant species in the chemical equilibrium. In addition, correlations of NH_3 total columns with
450 total columns of SO_2 and NO_x , and surface temperature were calculated. These results are shown in Table 9. The $\text{NH}_3/\text{NH}_4^+$ ratio is less than one for all sites except Porto Velho and Lauder, meaning that NH_3 prevails in the particulate phase. This suggests that the previously described model trends might be underestimated, as they were calculated for NH_3 in the gas phase. Most of the correlations with SO_2 and NO_x are very small, with a few exceptions. For Porto Velho, the $\text{NH}_3/\text{NH}_4^+$ ratio is 2.93 with $R = 0.91$ for SO_2 and $R = 0.77$ for NO_x . This suggests that for Porto Velho, the NH_3 trends are due to changes in
455 the emission sources rather than a reduction in the emissions of SO_2 and NO_x . The correlation coefficients between surface temperature and NH_3 total columns were > 0.5 only for Bremen, Garmisch, Zugspitze, and Jungfrauoch, suggesting that for these sites, temperature changes could have a greater impact on NH_3 concentrations than for other sites.

5 Conclusions

Using NH_3 total columns from ground-based FTIR spectrometers at 22 globally distributed sites and from a global CTM
460 (GCHP) and a reanalysis product (TCR-2), this work examined the variability and trends of atmospheric NH_3 . The FTIR time series revealed that NH_3 is present even in the most remote areas, and it can show significant enhancements, mainly due to biomass burning events. The seasonal patterns were similar across most sites, with total columns ranging between 2.66 and 6.92×10^{15} molec. cm^{-2} , and are in agreement with previous studies mainly using satellite data. The patterns are influenced by meteorological conditions such as temperature and rain, seasons of fertilizer application, and fire season. Overall, both
465 NH_3 simulations captured the seasonality of the FTIR measurements with some differences, mainly underestimations for the Northern and Southern Hemisphere sites and GCHP overestimations in the Arctic.

The diurnal variability of NH_3 total columns was shown for the first time for most of the FTIR sites, with total column variations between 0.20 and 6.74×10^{15} molec. cm^{-2} and large differences between sites. In general, the diurnal patterns differ significantly and depend on the characteristics of each site, as well as the influence of local sources such as vehicular
470 emissions and NH_3 transported from surrounding areas in the late afternoon. Both models struggled to capture the diurnal variability of the measurements, mainly due to the lack of diurnal variation in the anthropogenic emissions used in the NH_3 simulations.

Most NH_3 FTIR trends are positive and significant, ranging from 0.38 to 93×10^{13} molec. $\text{cm}^{-2} \text{ yr}^{-1}$ for all sites. The largest trends were for Mexico City, Boulder, and Toronto. The equivalent trends calculated using GCHP and the TCR-2
475 product were, in general, smaller for both GCHP and TCR-2. The trends using the full time series for GCHP are 10.11 (9.31 – 10.91) $\times 10^{13}$ molec. $\text{cm}^{-2} \text{ yr}^{-1}$ between 2003 and 2021 and 1.93 (1.11 – 2.75) $\times 10^{13}$ molec. $\text{cm}^{-2} \text{ yr}^{-1}$ for TCR-2 between 2005 and 2018. The drivers behind these trends depend on each location, and they involve a combination of changes in the



Table 9. GCHP $\text{NH}_3/\text{NH}_4^+$ total column ratio and correlation coefficients for NH_3 total columns vs. SO_2 and NO_x total columns and surface temperature between 2003 and 2021. Model values correspond to the nearest grid cell to each site. Surface temperature was vertically interpolated to station pressure levels to account for unresolved topography.

Station	Ratio	Correlation coefficient R for NH_3 with		
	$\text{NH}_3/\text{NH}_4^+$	SO_2	NO_x	Surface Temperature
Eureka	0.08	-0.03	0.13	0.07
Ny Alesund	0.08	0.00	0.07	-0.02
Thule	0.20	-0.17	0.48	0.38
St. Petersburg	0.14	-0.06	0.10	0.33
Bremen	0.57	0.02	-0.18	0.64
Garmisch	0.56	-0.15	-0.02	0.52
Zugspitze	0.56	-0.15	-0.02	0.59
Jungfraujoch	0.50	-0.13	-0.19	0.51
Toronto	0.22	-0.15	-0.04	0.40
Boulder	0.52	-0.03	0.39	0.49
Tsukuba	0.16	0.00	-0.06	0.12
Los Angeles	0.59	0.03	0.22	0.17
Hefei	0.23	-0.25	-0.25	0.25
Izana	0.19	0.09	0.30	0.19
Mexico City	0.24	0.40	0.09	0.47
Altzomoni	0.24	0.40	0.09	0.22
Paramaribo	0.92	0.01	-0.07	0.00
Porto Velho	2.93	0.91	0.77	0.41
Reunion St. Denis	0.21	-0.01	-0.14	-0.10
Reunion Maito	0.21	-0.01	-0.14	-0.06
Wollongong	0.55	0.37	0.63	0.26
Lauder	1.20	-0.10	0.17	0.16



NH₃, NO_x, and SO₂ emissions and changes in the meteorology. According to GCHP, NH₃ prevails in the particulate form NH₄⁺ at most sites, suggesting that the model trends presented in this work might be underestimated.

480 Overall, GCHP exhibited a generally better agreement with the FTIR observations than TCR-2, with the largest differences at the Arctic sites and Mexico City. Sensitivity tests regarding the a priori emissions were performed, finding that the differences in the emissions inventories used in GCHP and TCR-2 are not driving the larger underestimation observed in the TCR-2 product. The persistent negative bias in TCR-2 can be related to the lack of a bi-directional exchange flux scheme in its framework and to persistent positive model biases in sulfate and nitric acid. Further sensitivity tests and direct assimilation of NH₃ from
485 satellite observations are recommended to improve the TCR-2 NH₃ product.

NH₃ is a short-lived and highly variable atmospheric trace gas present in urban and remote areas, with consequences for the Earth's radiative forcing balance, ecosystems, and human health, including its role as PM_{2.5} precursor. Atmospheric NH₃ concentrations have been rising since the early 1900s with the introduction of synthetic fertilizers and they continue to increase globally. The lack of global regulation regarding NH₃ emissions, changes in meteorology, and emissions of other species such
490 as NO_x and SO₂ are some of the factors contributing to this. It is important to continue and expand the monitoring of NH₃ using ground-based and satellite instruments to extend the time series of this gas, analyze trends, and reduce uncertainties in the emissions and simulations. Such measurements could help inform the development of global regulations and policies that are needed to reduce the atmospheric concentration of NH₃ and its effects.

Appendix A: NH₃ Enhancements

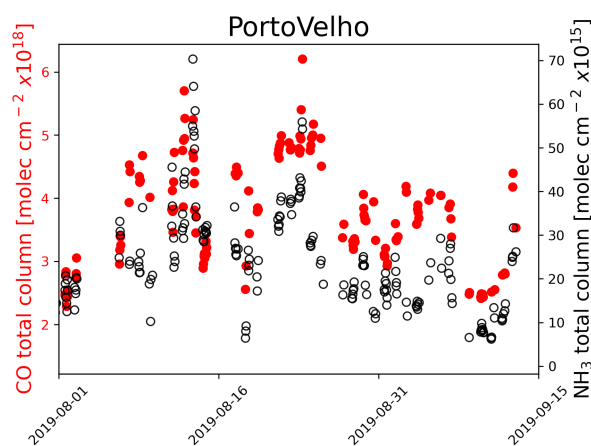


Figure A1. Comparison of NH₃ and CO total columns measured at Porto Velho between July and September 2019. Simultaneous enhancements can be observed, attributed to biomass burning events.

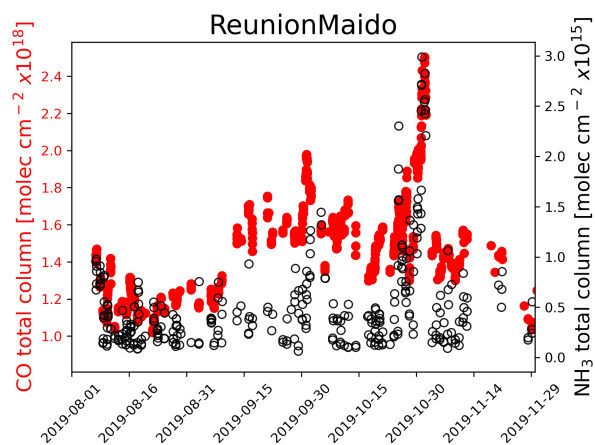


Figure A2. Comparison of NH_3 and CO total columns measured at Reunion Maido between August and November 2019. Simultaneous enhancements can be observed, attributed to biomass burning events.

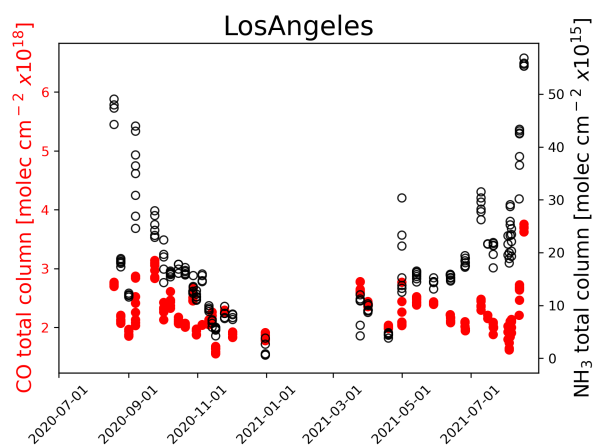


Figure A3. Comparison of NH_3 and CO total columns measured at Los Angeles between July 2020 and August 2021. Simultaneous enhancements can be observed, attributed to wildfires.

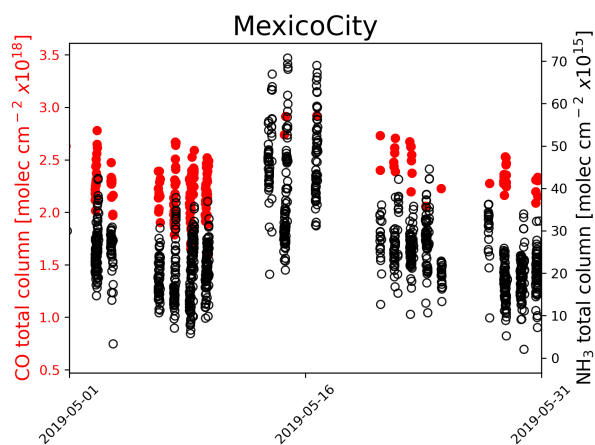


Figure A4. Comparison of NH₃ and CO total columns measured at Mexico City during May 2019. Simultaneous enhancements can be observed, attributed to biomass burning events.

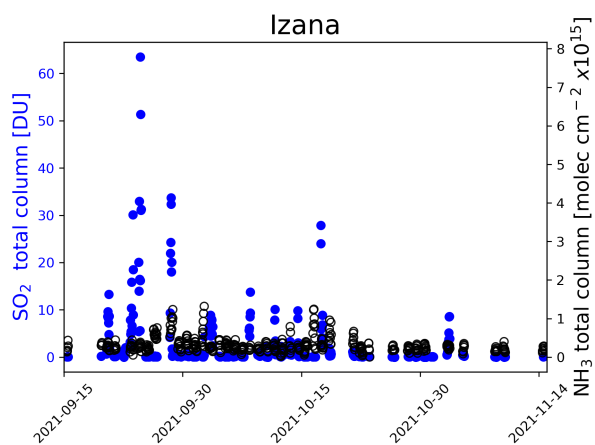


Figure A5. Comparison of NH₃ and SO₂ total columns measured at Izana between September and November 2021 during the Tarjogaite volcanic eruption that reached the Izana site. Simultaneous enhancements can be observed.



495 Appendix B: Model Comparisons

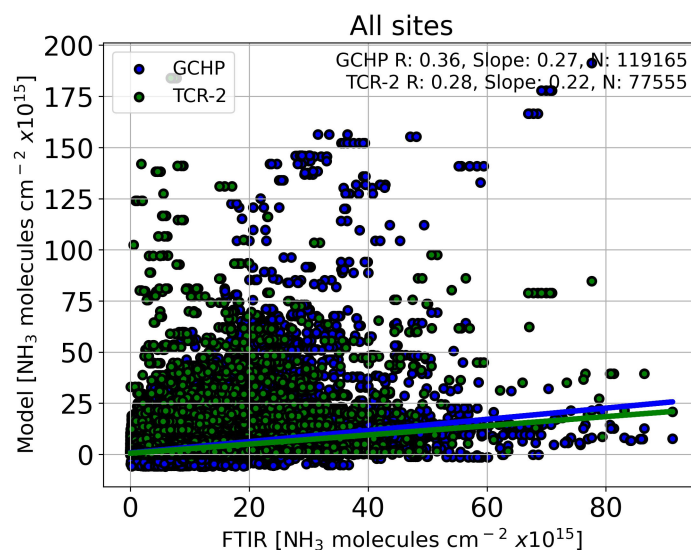


Figure B1. Scatter plot of GCHP and TCR-2 vs. FTIR temporally matched NH_3 total columns. The lines show the regression results for each model.

Data availability. The time series of FTIR NH_3 total columns are available on the Borealis repository at <https://doi.org/10.5683/SP3/9UV6KA> (Herrera Gutierrez et al., 2025). The TCR-2 products are available via <https://tes.jpl.nasa.gov/tes/chemical-reanalysis/> (Miyazaki et al., 2019). The GEOS-Chem High Performance model version 14.1.1 used in this work is freely available to the public via <https://doi.org/10.5281/zenodo.7600586> (GEOS-Chem, 2023).

500 *Author contributions.* BH, DBAJ, and KS developed the concept of the study. BH wrote the manuscript. KS reviewed all versions of the manuscript. BH, ED, MDM, OG, MG, JWH, NJ, EM, MK, IM, IO, MP, AP, DS, HS, WS, RS, GCT, CV, WW, and KS facilitated the FTIR retrievals. KM contributed to the TCR-2 product. TW, DBAJ, and BH contributed to the GCHP product. All co-authors provided critical review and comments throughout the different versions of the manuscript.

Competing interests. The authors declare that they have no conflict of interest.

505 *Disclaimer.* Language editing was assisted using Grammarly. The authors retain full responsibility for the manuscript content.

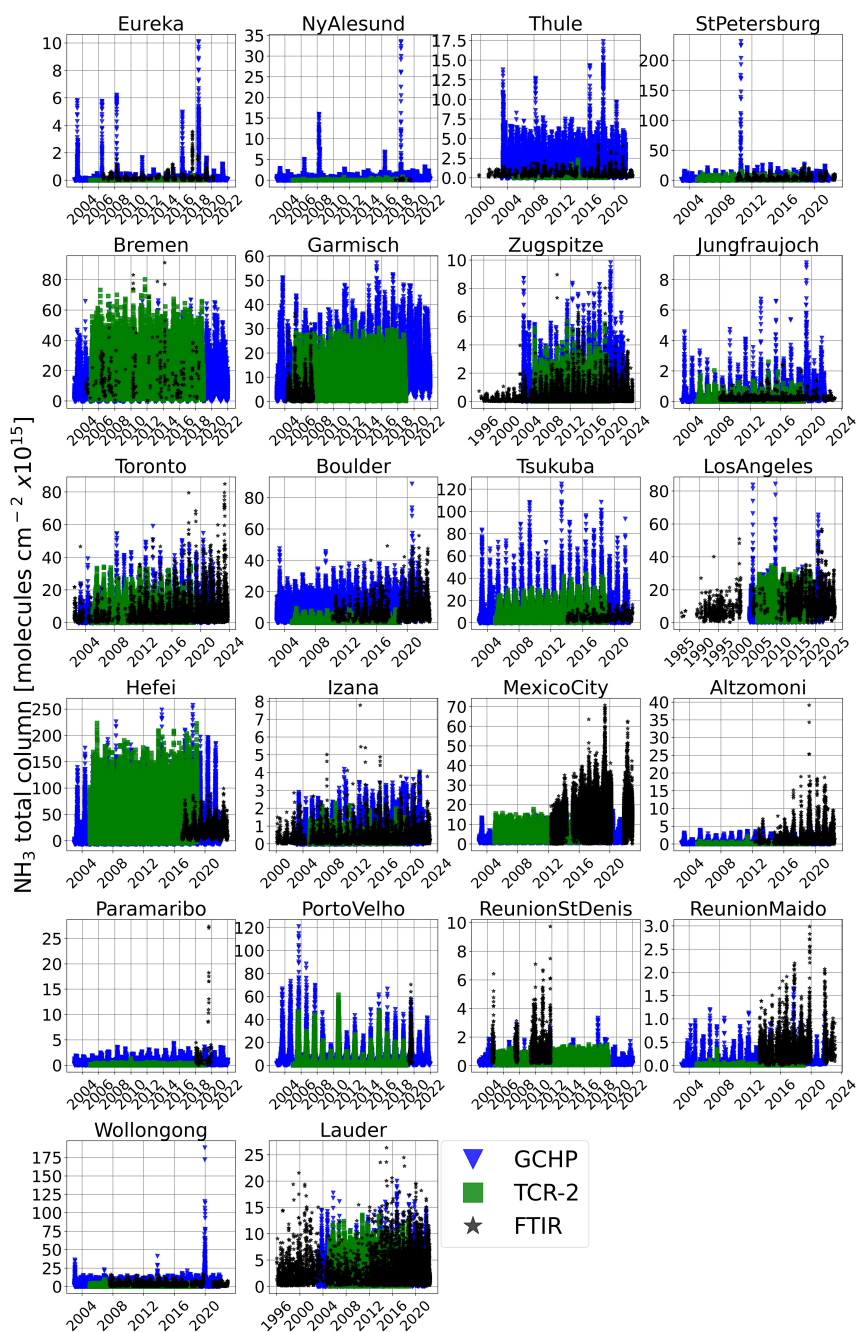


Figure B2. Comparison of the time series of NH_3 total columns from FTIR measurements (black) versus GCHP (blue) and TCR-2 (green) at the 22 FTIR sites.

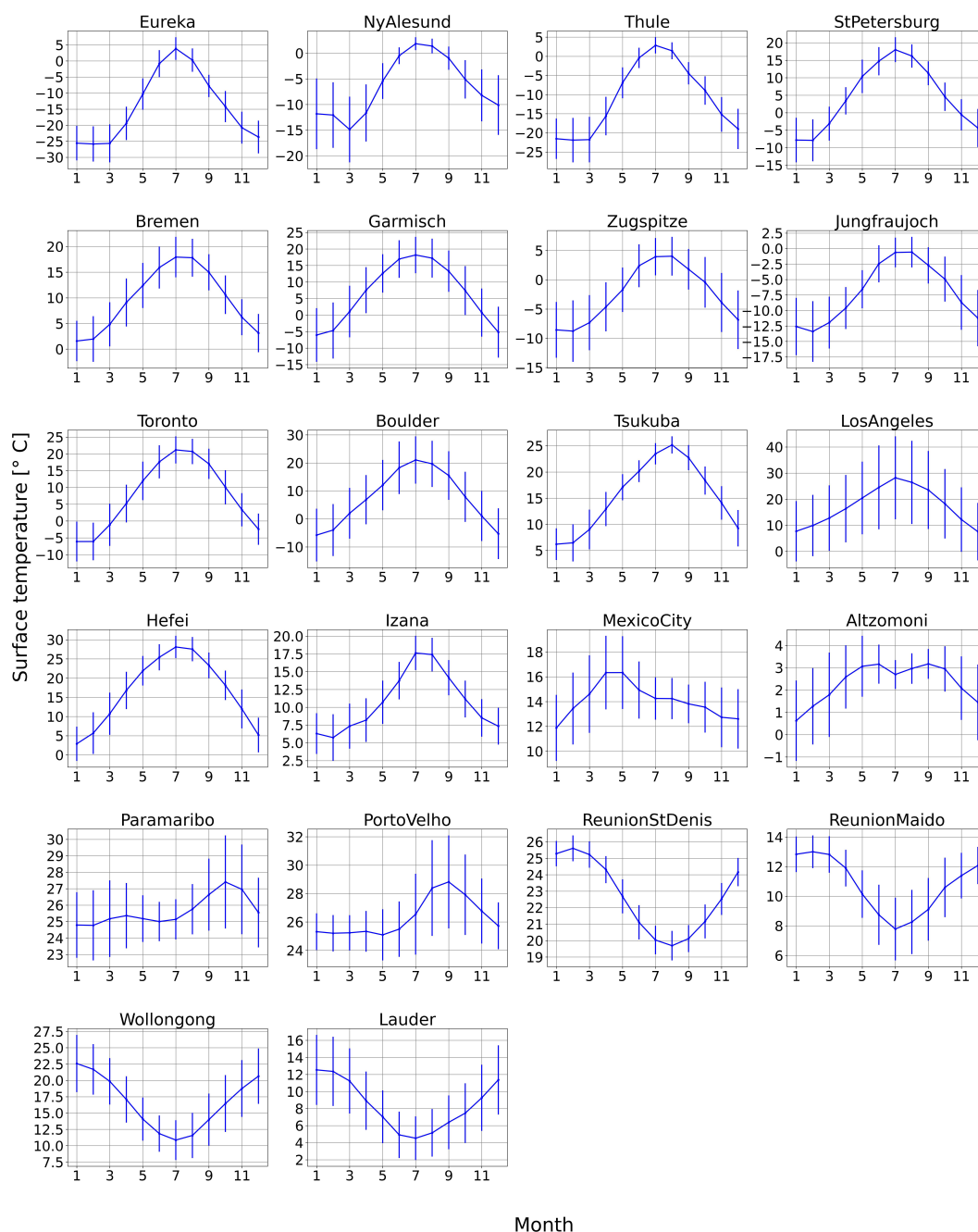


Figure B3. Monthly mean GCHP surface temperature at each FTIR site between 2003 and 2021. Vertical lines represent the standard deviation ($\pm 1\sigma$).



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